A STUDY OF STRONTIUM-90 LEVELS IN LOCAL MILK, TAPWATER, AND RAINWATER SAMPLES ST. JOHN'S, NEWFOUNDLAND

THOMAS BOYDE GRANDY

A STUDY OF STRONTIUM-90 LEVELS IN LOCAL MILK, TAPWATER, AND RAINWATER SAMPLES, ST. JOHN'S, NEWFOUNDLAND

by

Thomas Boyde Grandy, B.A. (Ed.), B.Sc.

Submitted in partial fulfilment of the
requirements for the degree of Master of Science
Memorial University of Newfoundland

September, 1962

This thesis has been examined and approved by:

R. K. Williams, B.Sc., Ph.D.

Assistant Professor

Memorial University of Newfoundland

K. G. McNeill, Ph.D.

Associate Professor

University of Toronto

TABLE OF CONTENTS

ABSTRACT

Strontium-90 activities in milk, rainwater and tapwater samples, collected in St. John's, Newfoundland, have been determined in the present investigation. Milk samples were collected weekly over two periods: January, 1961 to January, 1962 and July, 1962 to August, 1962. Rainwater and tapwater over the period May, 1962 to July, 1962 were studied. It was found that the strontium-90 levels in milk increased from a low of 2.4 to a high of 21.1 micromicrocuries per gram calcium over the experimental period. The activities in tapwater have also shown an increase from 0.25 micromicrocuries per litre in April, 1961 to 2.1 micromicrocuries per litre in July, 1962. Strontium-89 activities in rainwater and tapwater samples have also been computed.

During the course of the investigation a Philips low-level beta counter was calibrated with a standard strontium-90 source.

CHAPTER I

INTRODUCTION

During the year of 1960 the Physics Department of Memorial University of Newfoundland instituted the present project to determine the strontium-90 activities in environmental samples collected in Newfoundland. The importance of such measurements has been extensively discussed by various investigators.

Strontium-90 is one of the most abundant isotopes produced in fission bomb detonations. It is also chemically similar to calcium, which comprises the most of bone, and is consequently readily absorbed in the human body. Strontium-90, once absorbed, will continue to irradiate the bone for a considerably long time; the half-period of strontium-90 being 29.3 years (2). It can be seen then that the detection of radioactive strontium-90 in the human environment has considerable biological implications, especially, when frequent atomic weapon tests are carried out in the atmosphere.

The methods of strontium-90 analysis in the human environment have been extensively developed by previous investigators. Milk, which is one of the chief sources of dietary calcium, is generally analyzed to give some indication of the radioactive strontium-90 fallout and biological uptake (3). In addition to milk tapwater, rainwater, soil and air have been analyzed (5,7).

In the present investigation liquid milk samples were collected weekly over two periods; January, 1961 to January, 1962 and July 1962 to August, 1962. Rainwater and tapwater samples were also collected over the period May, 1962 to July, 1962.

Prior to strontium-90 activity measurements strontium was chemically extracted from each collected sample. A typical chemical analysis of milk is shown in Table 1 after the organic materials had been destroyed by ashing (10).

Table 1 Composition of Milk Ash

Compound	% by weight of whole milk
potassium oxide	0.175
sodium oxide	0.070
calcium oxide	0.140
magnesium oxide	0.017
ferric oxide	0.001
sulfur trioxide	0.027
phosphorus trioxide	0.170
chloride	0.100

It has been shown that the inorganic compounds listed in Table 1 could easily be separated from strontium (added to the milk as a carrier) by eluting them from an ion-exchange resin (3), for example, Dowex 50W - X12 cation exchange resin which was used in the present investigations. In the collected water samples, barium and cerium compounds were the only components which introduce some difficulty in the chemical separation of strontium (7). The separation of strontium and calcium in milk samples and the elimination of barium and cerium in water samples were therefore of prime importance in the chemical procedures employed in the investigation.

In counting strontium samples which are obtained after the chemical separation, the number of counts registered represents disintegrations in the sample of strontium-89 and strontium-90, which are the only long-lived

strontium isotopes present. It can be seen, therefore, that the strontium-90 activity of a collected sample cannot be measured directly. A study of the decay schemes of strontium-89 and strontium-90 gives the method of determination of strontium-90 activities. The decay schemes of strontium-89 and strontium-90 are given in Fig. 1.

It is noted that strontium-89 decays to the stable yttrium-89 and strontium-90 to the radioactive yttrium-90. Calculation shows that yttrium-90 reaches secular equilibrium with its parent, strontium-90, in approximately 17 days. The original strontium-90 activity can be found directly by chemically separating and counting the yttrium-90 after secular equilibrium is reached. In order to obtain absolute values of radioactivity in each sample the counting unit used was calibrated with a standard strontium-90 source and a standard yttrium-90 source.

The chemical procedures used in the present investigation are given in Chapter II followed by the description and standarization of the counting system in Chapter III. Final results and discussion are presented in Chapter IV.

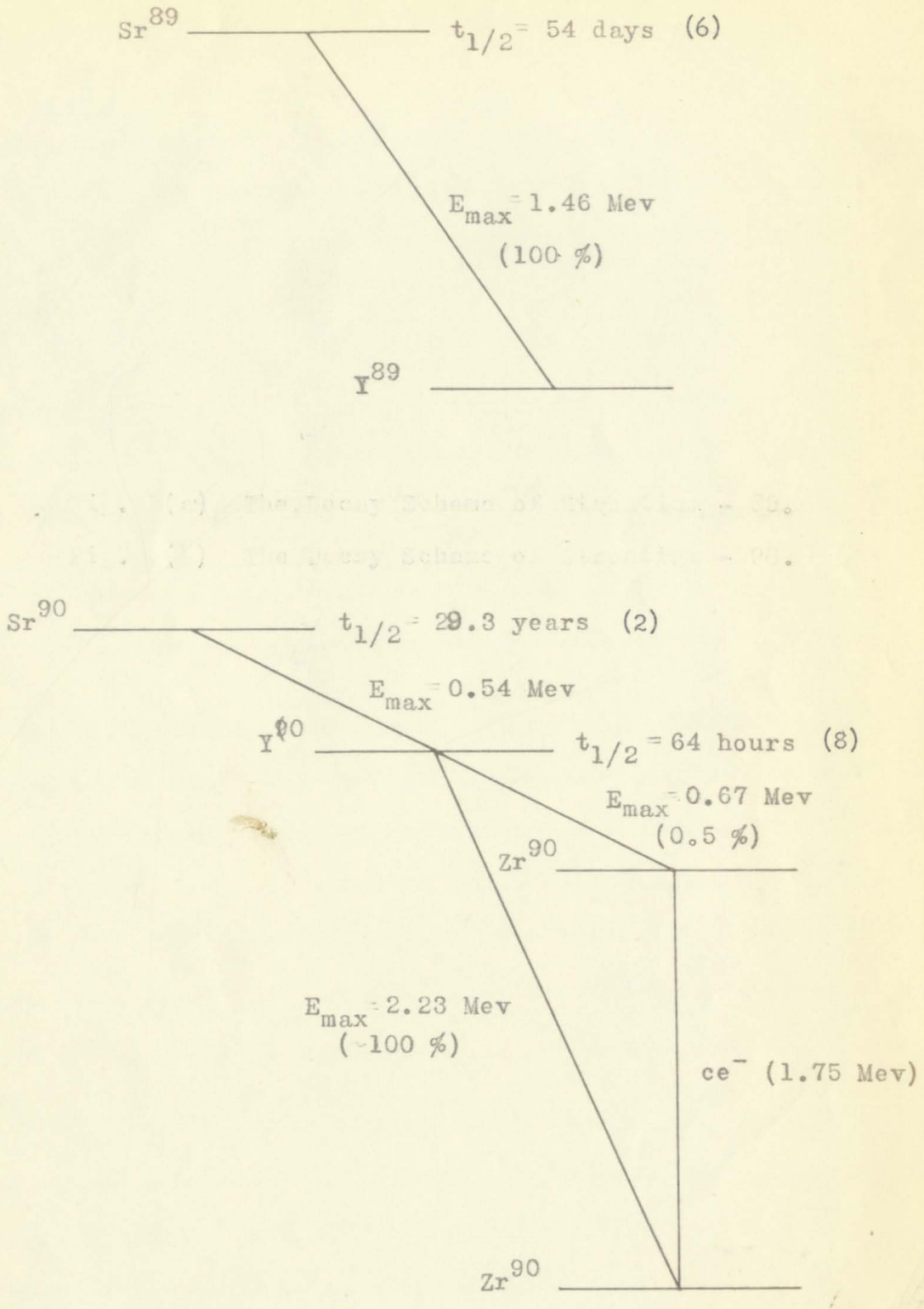


Fig. 1 - Decay Schemes of Strontium-89 and Strontium-90

CHAPTER II

CHEMICAL PROCEDURES FOR THE PREPARATION OF RADIOACTIVE STRONTIUM AND YTTRIUM SAMPLES

1. Strontium Separation from Milk

The determination of strontium-90 activities in liquid milk was carried out following the procedure previously developed by Atomic Energy of Canada Limited and The Department of Health and Welfare (3). Some alterations in the method were made in accordance with a paper by Osmond, Owers, Healy and Mead (7), specifically, as regard to the strontium purification. In Fig.2 the chemical procedure employed in this investigation is shown schematically.

The chemical procedure consists of ashing, Stage 1, separation of strontium from calcium, Stage 2, and purification of strontium, Stage 3.

Stage 1: Each milk sample, six quarts by volume, was reduced to its solid residue by slow evaporation of the water. The solid residue was then burned with a compressed air burner until the evolution of gas ceased (2-3 hours). Further ashing was carried out by heating in an electric muffle furnace at 700°C until the solid had a completely white appearance (approximately 12 hours). A sample of 0.5 gm of ash was reserved for the determination of calcium and 19 gm for the separation of strontium from calcium.

The percentage calcium content was determined for all milk ash samples since strontium-90 activities in milk are to be expressed in units of micromicrocuries per gm calcium. Quantitative analysis of calcium was made by means of its precipitation as the oxalate:

$$Ca^{++} + C_2O_4^{--} \longrightarrow CaC_2O_4 \downarrow$$

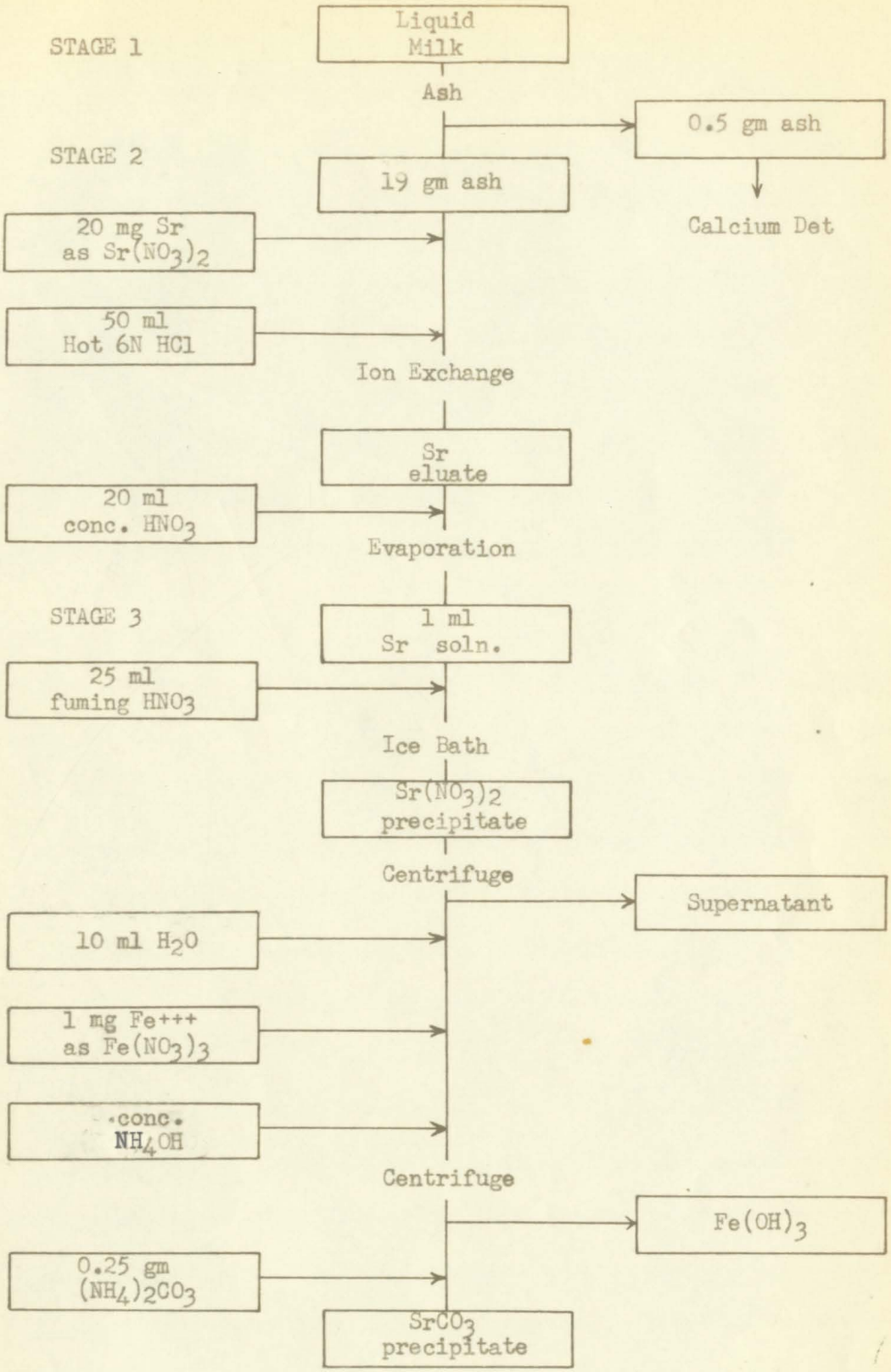


Fig. 2 - Chemical Procedure for Strontium separation From Milk

Brown and O'Donnell (4) outline a method for calcium analysis, following which the calcium oxide of the ashed milk was converted to calcium oxalate, washed free of soluble oxalate, treated with sulfuric acid and the liberated oxalic acid titrated with a standard permanganate solution.

Stage 2: One ml of strontium carrier solution containing 20 mg of strontium per ml was added to the milk ash from Stage 1. The carrier solution was prepared by dissolving 48.30 gm strontium nitrate in distilled water and diluting to a volume of 1000 ml. The sample was dissolved in 30-40 ml hot 6N HCL, diluted to 250 ml with distilled water, and passed through an ion-exchange column for separation of the calcium from strontium (3).

The ion-exchange column used in the analysis is shown in Fig.3 The ion-exchange column consisted of a resin column, A, and an influent tube, B, connected by a rubber stopper, C. The resin column, A, was constructed from a chromatographic tube 43 cm long and 2.0 cm inside diameter, equipped with a coarse porosity sintered glass filter, D. The resin column was surrounded by a glass cylinder, E, 40 cm long and 5 cm diameter and held in position by rubber stoppers, C, F. The temperature of the column A, was controlled by an electrical heater, H_1 . Dowex 50W-X12 cation exchange resin (200-400 mesh) was packed in the column, A, to a height of 30 cm above the sinter, D. The glass influent tube, B, 40 cm long and 1.1 mm inside diameter, was surrounded by a glass tube, G, 30 cm long and 3 cm diameter. The tube, G, contained water heated by the electrical heater, H_2, to insure uniform heating of the influent. The temperatures of E, G, were measured by means of thermometers inserted inside.

Prior to sample absorption approximately 150 ml 1.5 M ammonium

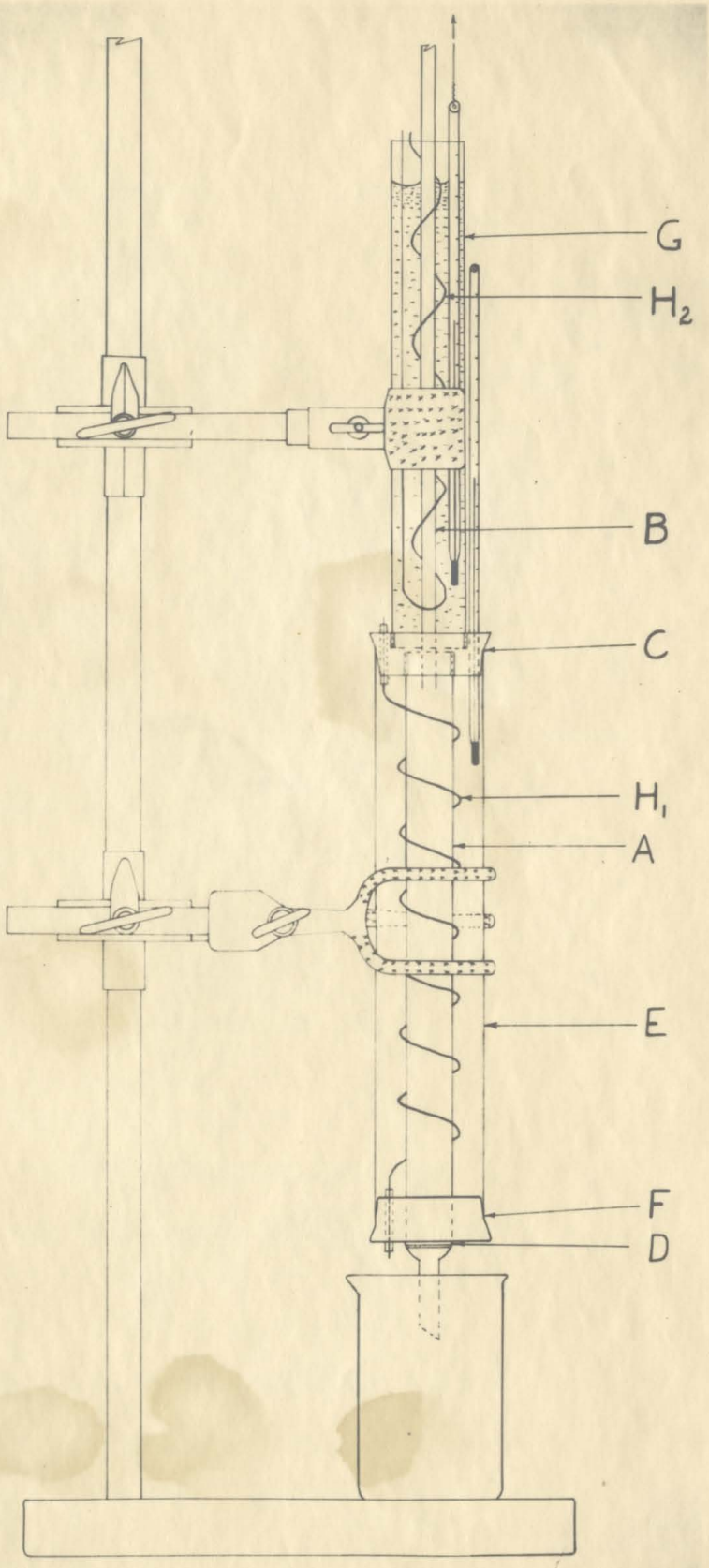


Fig. 3 - The Ion-exchange apparatus

lactate (pH 7) was allowed to run on the column. After about 30 ml had run through the heaters were turned on. For the sample adsorption step, which followed immediately, the temperatures were maintained at 78 ± 5°C for the influent tube and 65 ± 5°C for the resim column. The flow rate for sample adsorption was maintained at 4 to 5 ml per minute. Ammonium lactate with a molarity of 1.5 and pH 7 was used as an elutriant for calcium. Column temperatures and flow rates for the calcium elution were identical with those for sample adsorption. The end of the calcium band was determined by collecting small amounts of eluate in saturated oxalic acid and observing when calcium oxalate was no longer formed. Another 10-15 ml was collected to insure that all the calcium was off the column. Finally strontium was eluated with 350 ml 6N hydrochloric acid. Column temperatures were lowered to 70 ± 5°C and 55 ± 5°C, respectively, for the influent tube and resin column. At these lower temperatures the strontium elution flow rate was reduced to 2 to 3 ml per minute. The first 15-20 ml of the strontium eluate was discarded since it contained ammonium chloride and lactic acid which would impede the recovery of strontium. The first 15-20 ml contained only traces of strontium.

The column was regenerated by washing with 4N ammonium hydroxide until basic, then with 1.5 M ammonium lactate (pH 7) until pH of wash was 7.5.

20 ml concentrated nitric acid was added to the strontium eluate to destroy any remaining ammonium chloride. The total volume was then reduced to 1 ml by means of evaporation.

Stage 3: Strontium nitrate was precipitated in the evaporated strontium eluate by adding 25 ml fuming nitric acid and cooling in an ice bath for

30 minutes. This precipitate was centrifuged and the supernatent discarded. Any minute quantity of calcium that might have come along in the strontium eluate was considered to have been eliminated in the fuming nitric acid since calcium nitrate is much more soluble in the acid than strontium nitrate (5). The strontium nitrate was dissolved in 10-20 ml distilled water. Further purification of the above strontium solution was obtained by adding 1 mg iron carrier as ferric nitrate and precipitating ferric hydroxide with an excess of carbonate free ammonium hydroxide to act as a scavenging agent. The ferric hydroxide precipitate was centrifuged and the supernatant saved for the precipitation of strontium. After the precipitation of ferric hydroxide the purification procedure was considered to be complete. At this stage the time was noted for the purpose of estimating the amount of radioactive decay products present in the sample at any subsequent time. Strontium carbonate was precipitated as the final sample by adding 0.25 gm solid ammonium carbonate. This last precipitate was centrifuged and the supernatant discarded. The strontium carbonate was washed twice in 1-2 ml distilled water, transferred to a Whatman No. 42 filter paper, washed with alcohol, weighed as $SrCO_3$ and mounted on an aluminium planchet for counting as described in Chapter II, Section 4.

2. Strontium separation from rainwater and tapwater

For the determination of strontium-90 activities in water reference has been made to a previous investigation by Osmond, Owers, Healy and Mead (7). Again the procedure may be divided into three stages; removal of cerium, Stage 1, removal of barium, Stage 2 and purification of strontium, Stage 3. This chemical procedure is shown schematically in Fig. 4.

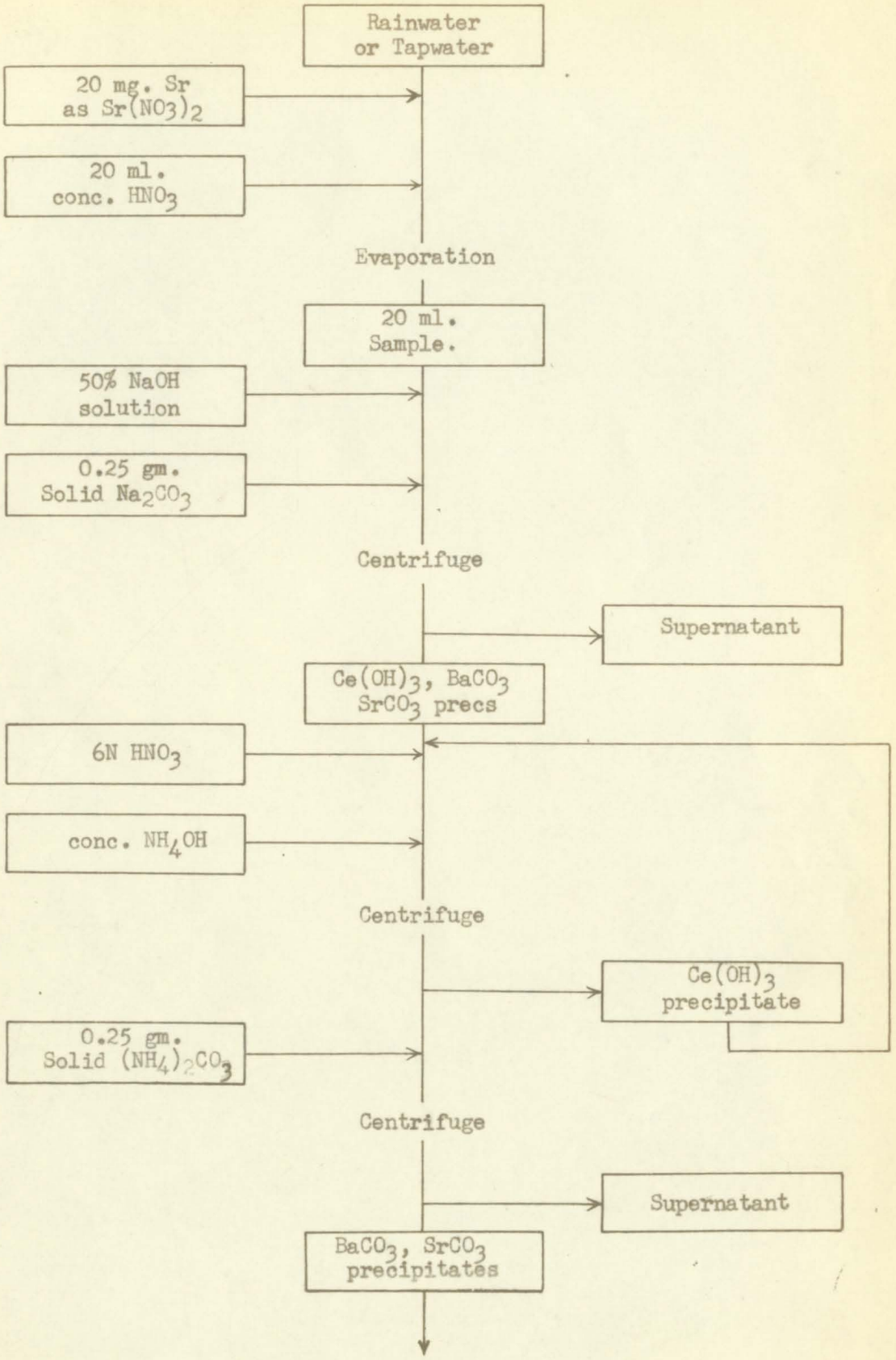


Fig. 4 - Chemical Procedure for Separation of Strontium From Rainwater and Tapwater.

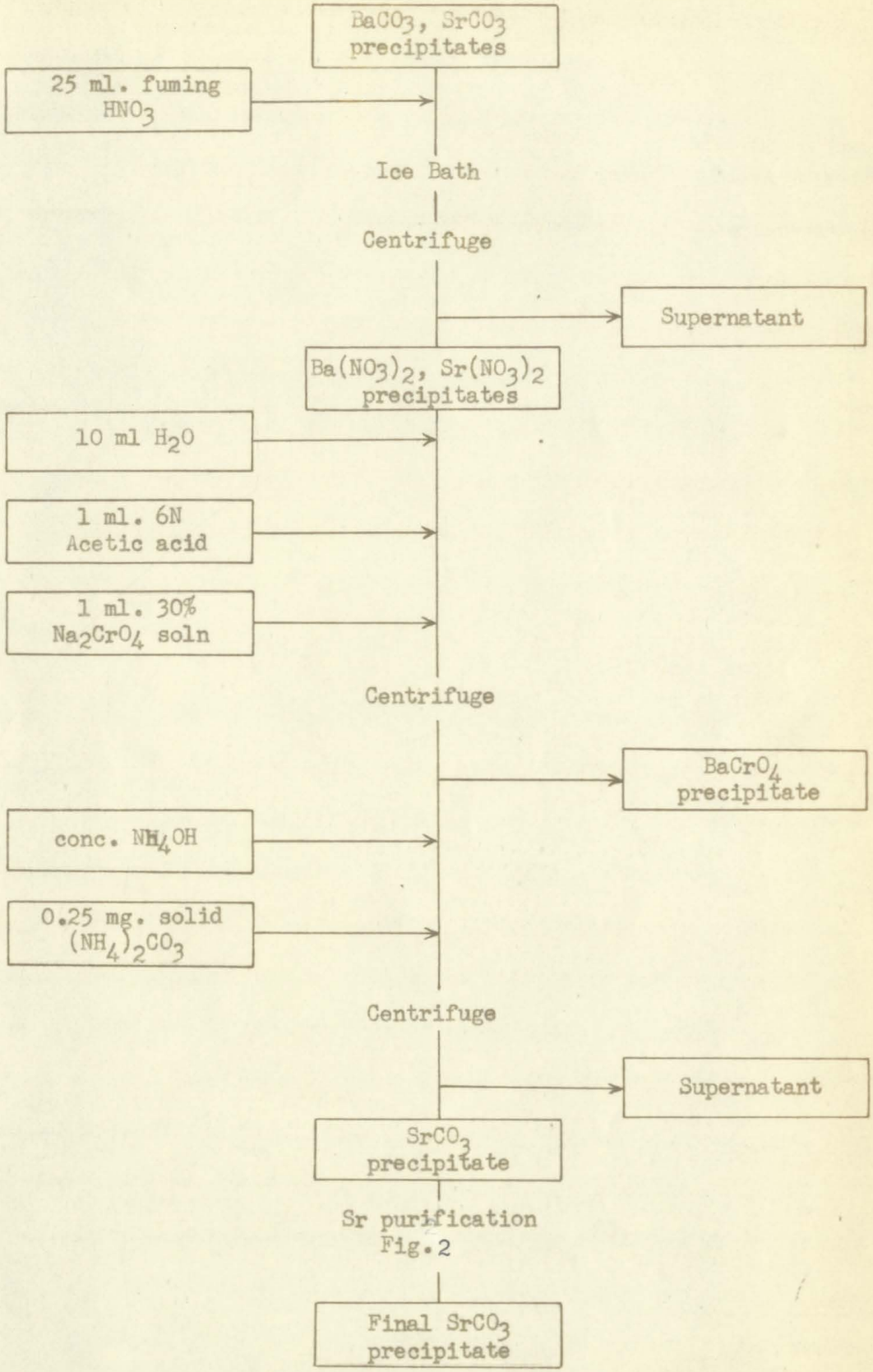


Fig. 4 (con't) - Chemical Procedure for Separation of Strontium From Rainwater and Tapwater

Stage 1: One ml of strontium carrier solution containing 20 mg of strontium per ml was added to each sample of rainwater and tapwater together with 20 ml concentrated nitric acid. The sample was then reduced in volume to 20 ml by evaporation and any residue filtered off. Cerium hydroxide was precipitated with sodium hydroxide and strontium carbonate with solid sodium carbonate. These precipitates were centrifuged and the supernatant discarded. The strontium carbonate and cerium hydroxide precipitates remaining were dissolved in 6N nitric acid and diluted to 30 ml. Cerium hydroxide was precipitated with an excess of carbonate free ammonium hydroxide and centrifuged. The supernatant was reserved for the separation of strontium. The precipitate was again dissolved in 6N nitric acid and cerium hydroxide reprecipitated as above in order to insure complete recovery of strontium. The cerium hydroxide precipitate was discarded.

Stage 2: Small amounts of barium, which might affect the accurate determination of strontium activities, were considered to be present in the final strontium solution from Stage 1 (7). These two elements were precipitated as the carbonates by adding 0.25 gm solid ammonium carbonate to this solution. The sample was then centrifuged and the supernatant discarded. Strontium nitrate and barium nitrate were precipitated by adding 25 ml fuming nitric acid and cooling in an ice bath for 30 minutes. The precipitates were centrifuged and the supernatant discarded. Strontium was finally separated from barium by first dissolving the nitrates in 10 ml water, then adding 1 ml 6N acetic acid, 2 ml 25% ammonium acetate solution and precipitating barium with 1 ml 30% sodium chromate solution. Strontium chromate was soluble in this buffered acetic acid solution. The sample was next centrifuged and strontium carbonate precipitated in the

supernatant with the addition of an excess of carbonate free ammonium hydroxide and 0.25 gm solid ammonium carbonate. The strontium carbonate was centrifuged and the supernatant discarded.

Stage 3: The final precipitate from Stage 2 was dissolved in 25 ml fuming nitric acid and the procedure carried on as indicated in Stage 3 for milk analysis.

Rainwater samples for which the chemical analysis has been described above were collected in two ways. One method utilized an ion-exchange collection system first developed by Aler and Edvarson (1), the other a funnel discharging directly into a four litre bottle. Both collectors are shown schematically in Fig. 5. The ion-exchange collector was constructed entirely of polyethylene and consisted of a funnel, A, an ion-exchange column, B, and a leveling device, C. The funnel and tapered tip, D, were threaded and could be replaced by standard bottle caps for shipment of the column by mail when sampling over a large area is to be done. The ion-exchange column had a length of 33 cm, an inside diameter of 2.5 cm and was packed to a height of 2.5 cm with Dowex 50X - X12 cation exchange resin (200-400 mesh). The funnels of both units measured 30.6 cm in diameter. Strontium collected with the ion-exchange column can be analysed by either eluting the column with 350 ml 6N hydrochloric acid or ashing the resin. In either event the strontium is subsequently subjected to the procedure described above for the separation of strontium from water.

Tapwater samples were collected in 8350 ml lots directly from the local water system. Flame spectrophotometric analysis showed that natural strontium in this volume was negligible in respect to the experimental error.

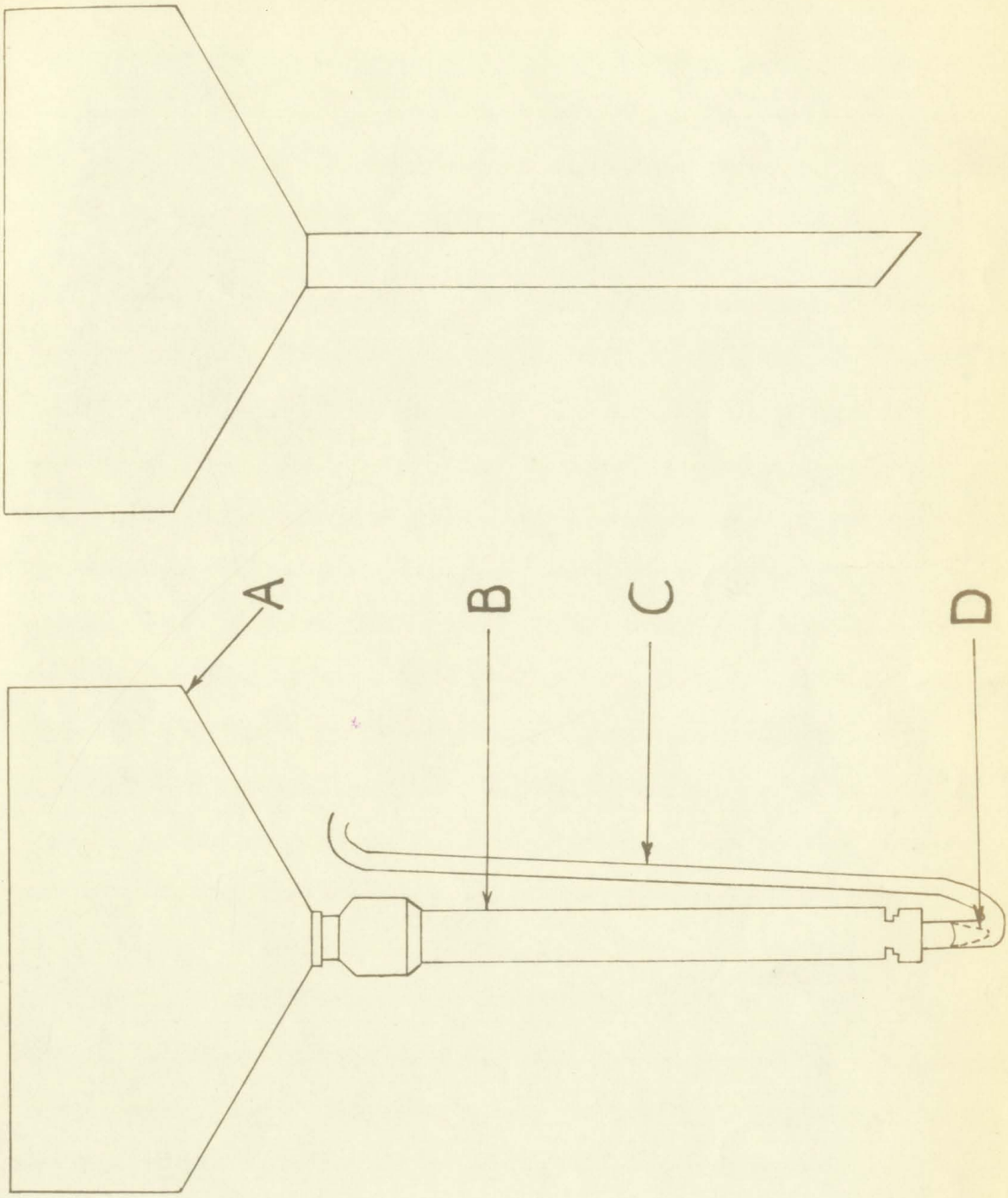


Fig. 5 - Rain Collectors

3. The Separation of Yttrium from Strontium Samples

Calculation shows that yttrium-90 reaches secular equilibrium with its daughter product strontium-90 in about seventeen days (approximately 7 yttrium-90 half-periods). Determination of the yttrium-90 content in the prepared strontium samples was therefore carried out at least seventeen days after the strontium purification.

The method of yttrium analysis employed in this investigation was adopted from a previous investigation by the Atomic Energy of Canada Limited and the Department of National Health and Welfare (3) and is shown schematically in Fig. 6. The strontium carbonate sample to be analyzed was scraped off the filter paper into a 50 ml centrifuge tube. The weights of the sample before and after transfer were checked in order to avoid unnecessary experimental error. 10 mg yttrium carrier, as yttrium nitrate, and 2-3 ml 6N hydrochloric acid were added to the sample and the tube placed in a hot water bath for fifteen minutes for yttrium exchange. Yttrium hydroxide was then precipitated with ammonium hydroxide, the sample centrifuged and the supernatant reserved for further strontium analysis, if necessary. The time of this first precipitation of yttrium hydroxide was noted for calculation after counting of the original yttrium-90 activity in the sample. The yttrium hydroxide precipitate was dissolved in 1 ml 6N hydrochloric acid, heated in a hot water bath for five minutes and yttrium hydroxide again precipitated, with ammonium hydroxide. The sample was next centrifuged and the supernatant discarded. This final yttrium hydroxide precipitate was once more dissolved in 1 ml 6N hydrochloric acid then diluted to 6 ml and heated in a water bath for five minutes. Yttrium oxalate was precipitated in this previous solution by adding 4 ml saturated oxalic acid with stirring and cooling in ice for just ten minutes. The yttrium oxalate precipitate was

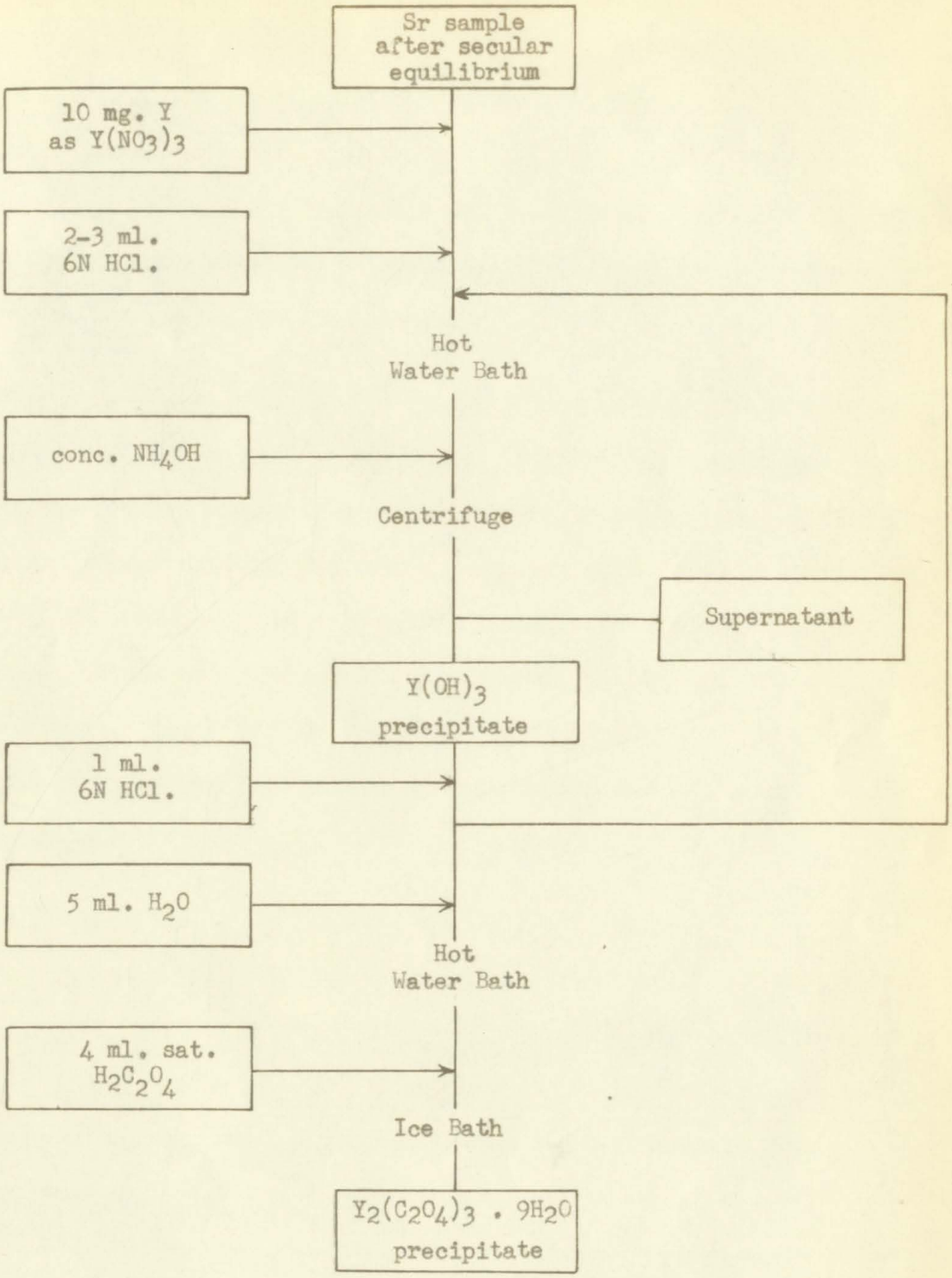


Fig. 6 - Chemical Procedure for Strontium-Yttrium Separation.

centrifuged and the supernatant discarded. The sample was washed with 3 ml distilled water, heated in a water bath for five minutes, centrifuged and the wash solution discarded. Another 2 ml of distilled water was added and the yttrium oxalate transferred to a Whatman No. 42 filter paper as described in the following section. Final washings were with 3-4 ml ethanol and ether. The sample was weighed as $Y_2(C_2O_4)_3 \cdot 9H_2O$ and mounted on an aluminium counting pan for counting.

The reference (3) used in the investigation gave the chemical formula of yttrium oxalate as $Y_2(C_2O_4)_3 \cdot 7H_2O$. Employing the drying procedure to be described in the following section it was found that the molecule was more highly hydrated than indicated in the above formula. In order to obtain the correct chemical formula an accurately known amount of yttrium nitrate was processed following the present experimental procedure, starting at the precipitation of yttrium oxalate. The final product was dried and weighed and the degree of hydration of the yttrium oxalate molecule determined.

4. Preparation of Radioactive Samples for Counting

Samples of strontium carbonate and yttrium oxalate obtained as described in the previous sections were prepared for counting by plating them onto a Whatman No. 42 filter paper. The filter paper had previously been cut into a disc 2.0 cm in diameter to fit an aluminium counting planchet. The chemical procedures for the preparation of strontium and yttrium samples required two washings in distilled water. The samples, while suspended in the second wash solution, were transferred to the filter paper with an eye-dropper. Fig. 7 shows the apparatus used in the sample plating procedure. At the top of the apparatus there was a coarse porosity sintered glass filter,

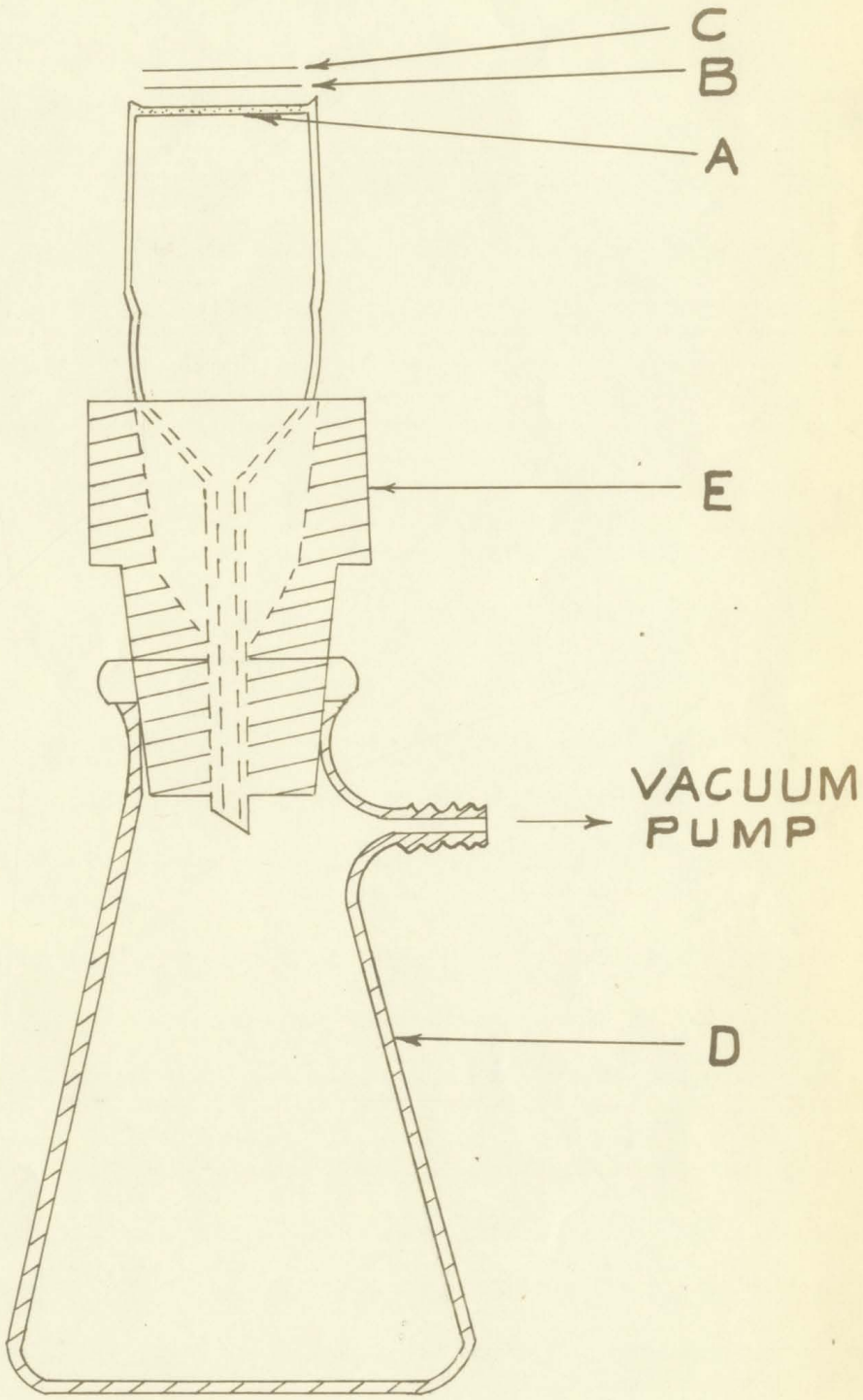


Fig. 7 - Sample Plating Apparatus

A, 2.0 cm in diameter; it supported a Whatman No. 40 filter paper, B, and the No. 42 filter paper, C. The filter paper, B, was used as a counterpoise to prevent direct contact of the sample with the sinter. The apparatus was supported by a vacuum flask, D, and a Walter crucible holder, E. The use of Whatman No. 42 filter paper enabled vacuum filtration to be employed during the plating. A high capacity vacuum pump was used to provide suction. The wash solution containing the suspended sample was transferred drop by drop to the filter paper, C; care was taken to insure a uniform deposit. Final sample washings were carried out with ethanol and ether. The use of these highly volatile wash liquids yielded a dry sample in a very short time. The transferred samples were dried in two ways; strontium carbonate samples under an infrared lamp and yttrium oxalate samples by running the vacuum pump at full with the sample still on the sinter. Drying by suction was advantageous in that the samples were held rigid against the sinter thus preventing curling of the filter paper and cracking of the sample. This method was necessary for the yttrium oxalate samples since they were highly hydrated and would soon have decomposed under an infrared lamp.

The method of plating by filtering unto a filter paper is recommended since it ordinarily yielded very uniform deposits, was relatively fast, showed little transferal loss and used a minimum of liquid in washing.

Samples were weighed immediately following drying and then attached to an aluminium counting pan with a small amount of Goodyear Pliobond cement. Care was taken to limit the amount of cement used to a minimum. This was necessary since cement might otherwise have soaked up into the sample and absorbed some of the radiation.

CHAPTER III

THE COUNTING APPARATUS

1. Description of the Apparatus

Sample activities were measured with a Philips anti-coincidence low background beta counter. Fig. 8 shows the main features of the Philips 4127/02 Anti-coincidence Lead Castle which housed the counting assembly. The unit consisted of a shield, A, mounted on a tripod, B, in which the counting tubes were located. The shield, A, was a hollow cylinder rounded off at the top into a hemispherical shape. It was constructed of iron 10 cm thick lined with very old lead 3 cm thick. Old lead was used since it was supposed to be free from any radioactivity.

A Philips 18515 Geiger tube, C, and a Philips 18518 guard tube, D, were situated inside the cavity of the shield, A. The beta counter, C, 37 mm in length, employed a mica window, diameter 34 mm and thickness 1.5 mg/cm^2. Its cathode was made short to decrease the sensitivity of the tube to gamma radiation. The guard tube, D, surrounded all but the end window of the Geiger tube, C. The guard tube was a Geiger-Muller tube of special construction; its electrodes consisted of two co-axial cylinders rounded off at the top with concentric hemispheres. The beta counter, C, was fitted, with the window facing downward, at the top of an opening, E, in the shield. Through the opening, E, the specimen to be measured was brought up to a position close to the window of the beta counter.

Each radioactive sample was placed at the top of a specimen support, F; the sample could be moved into and out of the opening, E, by means of an

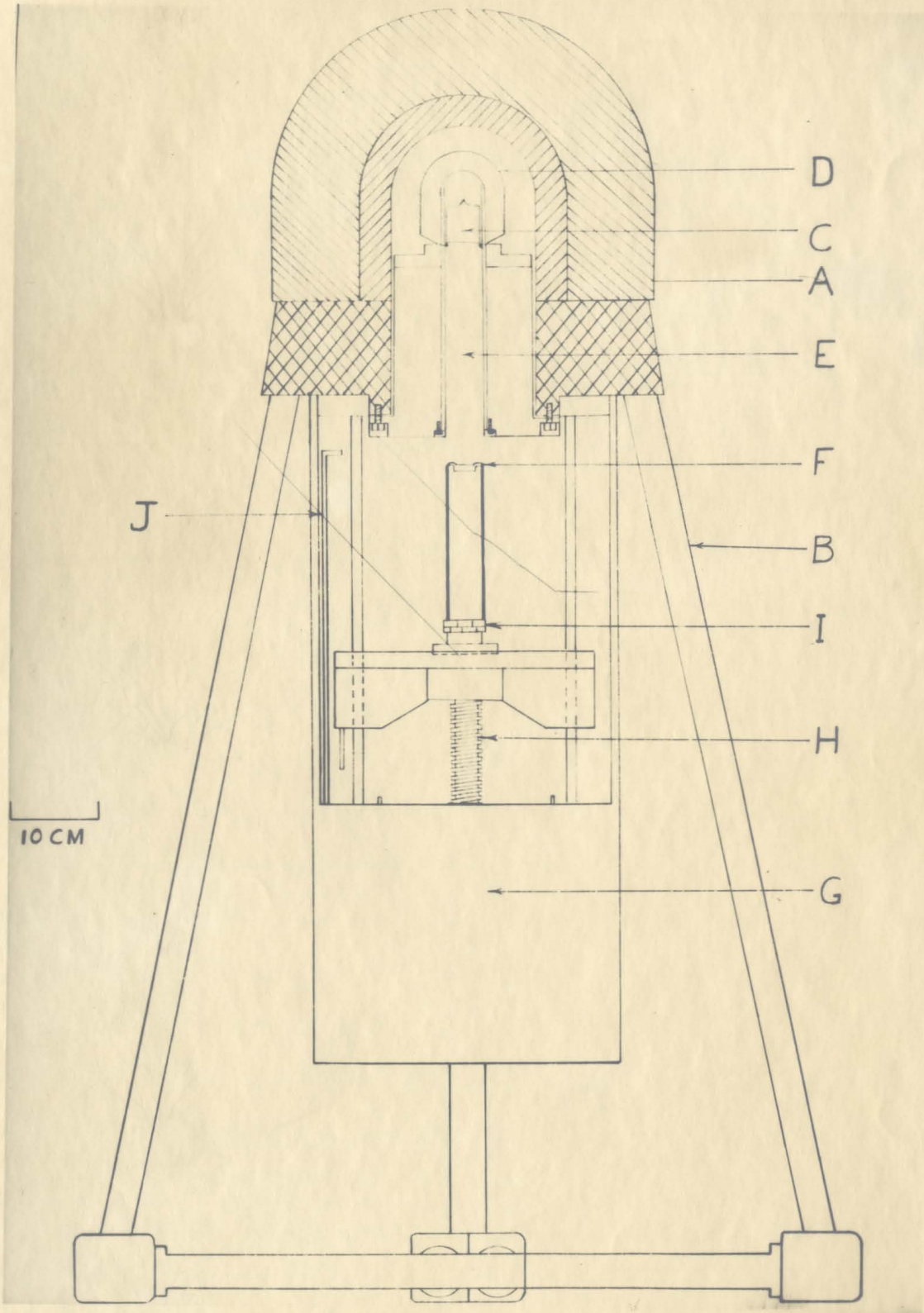


Fig. 8 - Philips' Anti-coincidence Lead Castle

electric motor situated inside the casing, G, via a worm spindle, H. In order to insure that every sample was brought up to the same position with respect to the end window of the beta counter, C, an adjustment was made to the stops, I, at the beginning of the project. The stops, I, were threaded rings and could be locked into any desired position. The ascent of the specimen support, F, was terminated by means of an automatic switch, J. Final adjustment of the positioning was made by means of a shorting-out switch located at the bottom of the apparatus.

Fig. 9 shows the electronic units employed in the counting system. High potentials were supplied to the counting tubes by two Philips 4022 High Voltage and Amplifier units; the operating voltage for the beta counter was 540 volts and for the guard tube 1060 volts. The outputs of the beta counter and guard tube were fed to a Philips 4092 Anti-coincidence unit; pulses from the beta counter were delayed 3 microseconds. Output pulses from the anti-coincidence unit were fed to two Philips 4032 Scalars. The first scalar registered pulses reaching it from the beta counter; the anti-coincidence circuit served to block pulses to this scalar for a predetermined time after the guard tube was triggered. Thus radiations which originated outside the apparatus, penetrated the shield and caused pulses in both tubes would not be registered by the beta counter scalar. This arrangement served to reduce the background counting rate in the beta counting system. The second scalar registered pulses from the guard tube through the blocking channel.

The anti-coincidence unit could be set to block the beta counting channel for times adjustable from 10 to 3000 microseconds. It was necessary to set this anti-coincidence dead time at some time greater than the dead time of the guard tube. The dead time of the guard tube was 1000 micro-

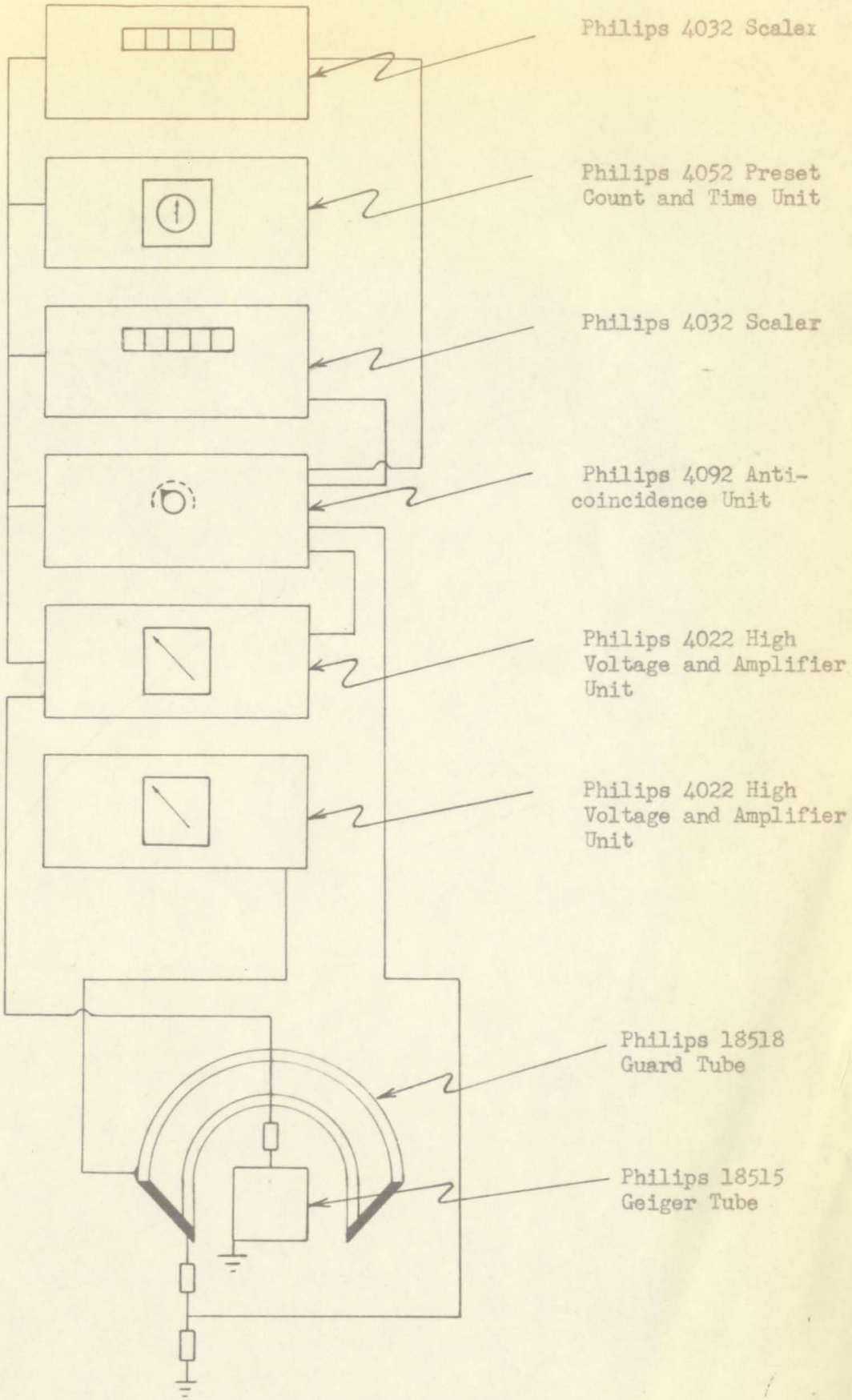


Fig. 9 - Electronic Units of the Counting System.

seconds, consequently the anti-coincidence dead time was adjusted to the closest setting provided by the unit; namely, 3000 microseconds. Imposing a dead time on the beta counting channel required a correction to be made in the observed counting rates. This correction was of the first order; that is, as regard to the experimental error it was necessary to correct only for the counting time lost because of the dead time.

Counting times were measured by a Philips 4052 Preset Count and Time unit which was controlled by the beta counter scalar. The clock employed in the unit was spring-driven and thus independent of mains frequency variations. It had a tolerance of 0.2 second.

Measurements of the counting tube characteristics were made at the beginning and termination of the experimental work presented in this investigation. The results of these measurements are shown in the following table.

TABLE 2 MEASURED CHATACTERISTICS OF THE COUNTING TUBES

TYPE OF TUBE	STARTING VOLTAGE (VOLTS)		PLATEAU LENGTH (VOLTS)		SLOPE OF PLATEAU %/100 VOLTS	
	Beginning of exp.	end of exp.	Beginning of exp.	end of exp.	Beginning of exp.	end of exp.
Geiger tube 18515 (operating voltage 540 volts)	340	340	475 to 650	475 to 650	1.0	1.0
Guard tube 18518 (operating voltage 1060 volts)	670	760	850 to 1250	980 to 1220	0.5	0.5

It can be seen from the table that the Geiger tube characteristics remained the same over the experimental period. Small changes were observed in the guard tube characteristics as regard to the length of the plateau and the starting voltage; however, this did not affect the experimental observations since the plateau slope remained constant and the operating voltage of the tube was fixed well within that range of the plateau which remained constant.

The voltage fluctuations of the high voltage supply units used in this investigation were given by the manufacturer as less than 0.01% for 1% mains voltage fluctuations. Output voltages were guaranteed to drift less than 0.1% during 10 hours.

It should be noted that during the entire experimental period the electronic units described above were not shut off.

Table 3 shows a number of background counting rates measured with the above apparatus and taken during the course of the experimental work.

2. Standarization of the Counting Unit

The efficiencies of the counting system with respect to strontium-90 and yttrium-90 samples were determined with a standard strontium-90 source obtained from Amersham, England. This standard had a strontium-90 activity of 15.4 microcuries on August 24th, 1961. The standard was received in the chemical form of strontium nitrate dissolved in 100 ml 0.01N nitric acid. The effective standard deviation in the calibration of the source was quoted as being approximately $\pm$ 2%.

A second standard solution of lower activity was prepared by diluting

TABLE 3 - BACKGROUND COUNTING

No.	Duration of Count (min)	Total Geiger Tube Count	Geiger Tube Counting Rate (c/m)	Total Guard Tube Count	Guard Tube Counting Rate (c/m)
1	1378	2648	1.93	89670	65.1
2	208	393	1.90	13659	65.7
3	670	1274	1.91	44042	65.7
4	1053	2002	1.91	68904	65.4
5	1021	1891	1.86	66444	65.1
6	510	1029	2.02	33284	65.3
7	217	398	1.84	14135	65.1
8	731	1359	1.87	48075	65.8
9	1064	2042	1.93	69368	65.2
10	1042	1941	1.87	67433	64.7
11	325	623	1.93	21150	65.1
12	1711	3293	1.93	111273	65.0
13	1140	2211	1.95	73810	64.7

The mean background counting rate was taken to be

1.9 ± 0.1 c/m

1 ml of the original to 1000 ml in 0.01N nitric acid. The strontium-90 activity of this standard solution was computed to be 151 micromicrocuries per ml when the calibration procedure commenced. Standard samples with activities ranging from 4.3 to 151 micromicrocuries were prepared for the standardization of the counting unit by further dilutions.

Strontium-90 solid sources were prepared as strontium carbonate by employing a procedure identical to that of the purification of strontium outlined for milk analysis (Stage 3). Similarly yttrium-90 solid sources were prepared as yttrium oxalate in accordance with the strontium - yttrium separation procedure outlined in Chapter II Section 3. These methods of preparation were used so that samples employed in the standardization had exactly the same form as the actual samples. In order to further insure that the yttrium-90 standard sources were strictly the same as the actual samples, strontium-90 solid sources were prepared and counted, then allowed to decay until yttrium-90 reached secular equilibrium with strontium-90, followed by the chemical extraction and counting of the radioactive yttrium.

In all cases 20 mg of strontium carrier and 10 mg of yttrium carrier were added to the standard samples. Strontium-90 solid sources were weighed as $SrCO_3$ and yttrium-90 sources as $Y_2(C_2O_4)_3 \cdot 9H_2O$; strontium and yttrium yields were computed for each and reported as a percentage. Specific activities were found by applying the appropriate percentage yield.

The counting efficiency for yttrium-90 was found from the ratio of the number of counts, Ny, observed over the counting period to the expected total number of disintegrations, Dy, in the sample for the same

period. The yttrium-90 activity, A(t), of the sample at any time, t, is given by

$$A(t) = Aoe^{-\lambda t}$$

where Ao = activity of the yttrium-90 sample at the times of separation of strontium from yttrium; disintegrations per minute

λ = disintegration constant for yttrium-90.

Radioactive source measurements were carried out over some period (t_2-t_1) after the separation of strontium from yttrium; t_1 and t_2 were measured from the time when the yttrium-90 activity was Ao. The total number of disintegrations in the sample over this period is given by

$$\int_{t_1}^{t_2} A(t)\,dt = \int_{t_1}^{t_2} Aoe^{-\lambda t}\,dt$$

$$= \frac{Ao}{\lambda}\left(e^{-\lambda t_1} - e^{-\lambda t_2}\right)$$

If Ny represents the total number of counts registered by the counting unit over the period (t_2-t_1), corrected for background and anti-coincidence dead time, then the counting efficiency for yttrium-90 is

$$Cy = \frac{Ny\lambda}{Ao\left(e^{-\lambda t_1} - e^{-\lambda t_2}\right)}$$

As for yttrium-90, the strontium-90 counting efficiency was found by computing the number of disintegrations of strontium-90 in the sample over the counting period and comparing this with the count observed due to strontium-90. The total count observed was due to strontium-90 and the increasing build up of yttrium-90. To find the number of counts due to strontium-90 it was necessary to first compute the contribution due to yttrium-90.

The total yttrium-90 activity, $A_2(t)$, in the strontium-90 sample at any time, t, is given by

$$A_2(t) = A_1^o (1 - e^{-\lambda_2 t})$$

where A_1^o = initial activity of strontium-90 (assumed constant over the experimental period); disintegrations per minute.

λ_2 = disintegration constant for yttrium-90.

The total number of disintegrations of yttrium-90 that occur after the purification of strontium over the counting period (t_2-t_1) is given by

$$\int_{t_1}^{t_2} A_2(t)\,dt = \int_{t_1}^{t_2} A_1^o (1 - e^{-\lambda_2 t})\,dt$$

$$= A_1^o \left[(t_2-t_1) - \frac{1}{\lambda_2} (e^{-\lambda_2 t_1} - e^{-\lambda_2 t_2}) \right]$$

where t_1 and t_2 are measured from the time of precipitation of ferric hydroxide in the purification procedure. The counting efficiency for yttrium-90 has previously been designated Cy. Thus the number of counts registered owing to the presence of yttrium-90 will be

$$Ny = Cy\, A_1^o \left[(t_2-t_1) - \frac{1}{\lambda_2} (e^{-\lambda_2 t_1} - e^{-\lambda_2 t_2}) \right]$$

If N_{obs} = total observed count corrected for background and anti-coincidence dead time, then the number of counts due to strontium-90, N_{Sr}, is

$$N_{Sr} = N_{obs} - Ny$$

The counting efficiency for strontium-90 is given as

$$C_{Sr} = \frac{N_{Sr}}{A_1^o (t_2-t_1)}$$

Tables 4 and 5 give the results for the standardization of the counting unit for yttrium-90 and strontium-90. Results showed no significant correlation between the counting efficiences and the ratio of source strength to added carrier weight of constant amount. In view of this the counting efficiency for each individual nuclide was assumed to be constant over the range of activities studied. The final result for each series of measurements was found by taking a weighted mean of total. Each individual result was given a weight proportional to the reciprocal of the square of its error. The weighted mean value $\bar{X}$ was computed as

$$\bar{X} = \frac{\sum_{n} \omega_i x_i}{\sum_{n} \omega_i}$$

where x_i = a measured value

ω_i = the weight assigned the value x_i

n = the number of individual determinations.

The standard deviation in the mean of weighted values was found as

$$\bar{X} = \left[\frac{\sum_{n} \omega_i (x_i - \bar{X})^2}{(n-1) \sum_{n} \omega_i} \right]^{\frac{1}{2}}$$

The yttrium-90 and strontium-90 counting efficiencies were found to be 40.5 $\pm$ 0.3% and 28.9 $\pm$ 0.3%, respectively. Since no strontium-89 standard was available an average of the above values was taken as the strontium-89 counting efficiency, namely, 35 $\pm$ 6%. The value was computed in this way since the strontium-89 beta end point energy is intermediate between those of strontium-90 and yttrium-90. These values were subsequently used to calculate final activities in the actual samples.

TABLE 4 - YTTRIUM-90 COUNTING EFFICIENCY

Sample No.	Original Activity (d/m)	Final Yield of Yttrium (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected Initial Counting Rate (c/m)	Counting Effeciency (%)
			Counting began t_1	Duration of count (t_2-t_1)				
1	302	96.1	87	39	4510	2632	122.6	40.6 ± 1.6
2	295	99.1	128	55	6383	3589	117.9	40.0 ± 1.5
3	170	98.8	243	57	3716	3701	67.3	39.6 ± 1.6
4	162	96.1	303	24	1475	1567	63.9	39.4 ± 1.8
5	85.0	92.0	291	114	3612	7517	34.7	40.8 ± 1.8
6	85.0	91.1	181	108	3518	7104	35.3	41.5 ± 1.9
7	51.9	90.8	112	110	2264	7191	21.3	41.0 ± 1.6
8	51.9	91.4	2023	223	3453	14648	21.8	42.0 ± 1.9
9	35.3	100.0	571	180	2648	11752	14.4	40.8 ± 1.7
10	34.2	99.7	1280	210	2707	13928	14.1	41.2 ± 1.7
11	16.9	96.9	249	349	2772	23218	6.75	39.9 ± 2.0
12	16.2	98.3	715	549	3848	35586	6.23	38.5 ± 1.9
13	8.4	100.0	1490	722	3032	47309	3.22	38.3 ± 4.2

TABLE 5 - STRONTIUM-90 COUNTING EFFICIENCY

Sample No.	Original Activity Added (d/m)	Final Yield of Strontium (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected Initial Counting Rate (c/m)	Counting Efficiency (%)
			Counting began t_1	Duration of Count (t_2-t_1)				
1	335	90.0	45	33	3054	2174	99.4	29.5 ± 1.2
2	335	87.0	88	29	2604	1875	99.7	29.8 ± 1.3
3	190	91.0	79	172	8821	11322	52.3	27.5 ± 1.2
4	190	88.5	300	592	33164	38396	53.6	28.2 ± 1.2
5	95.0	96.0	62	43	1269	2842	28.0	29.5 ± 1.4
6	95.0	94.0	31	28	722	1805	25.5	26.8 ± 2.1
7	95.0	96.0	219	74	2306	4876	29.3	30.3 ± 1.4
8	57.0	93.0	1309	101	2221	6680	16.8	29.5 ± 1.3
9	57.0	95.0	421	885	18485	57674	16.8	29.5 ± 1.2
10	38.0	94.5	51	66	795	4386	10.6	27.9 ± 1.6
11	38.0	93.0	120	268	3320	17587	10.6	27.9 ± 1.2
12	9.5	90.0	1405	139	723	9150	2.78	29.3 ± 3.2

CHAPTER IV

RESULTS AND DISCUSSION

A selection of data sheets, which were used in computing strontium-90 and strontium-89 activities in the various samples, is given in Tables 6 to 13.

As has been mentioned in the introduction, the strontium-90 of a particular sample must be computed from the activity of its daughter product, yttrium-90; this procedure is necessary because of the presence of strontium-89. For example, Table 6 gives the results for the yttrium-90 analysis of the milk samples. In this table, the following points should be noted:

(1) The third column gives the chemical yield of yttrium on the basis of 10 mg of yttrium carrier added.

(2) The fourth and fifth columns give the counting times; t_1 represents the time counting began and t_2 the time counting ended; both t_1 and t_2 are measured from the time of the first yttrium hydroxide precipitation.

(3) The total number of observed counts for the geiger tube and guard tube are shown in the sixth and seventh columns. The geiger tube count has been included to give some idea of the counting errors and the guard tube count to indicate anti-coincidence dead time corrections.

(4) The eighth column gives the initial yttrium-90 activity, or alternatively, the strontium-90 activity, of the samples.

(5) The final column gives the strontium-90 (yttrium-90) activity reduced to units of micromicrocuries per gm calcium. Final activities of yttrium-90 are corrected for decay before and during counting by

TABLE 6 - CALCULATION SHEET FOR DETERMINATION OF YTTRIUM-90 ACTIVITY IN MILK SAMPLES

Sample No.	Final Yield of Sr (%)	Final Yield of Y (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected initial Y^{90} activity of sample (uuc)	Activity of sample (uuc/gm Ca)
			t_1	(t_2-t_1)				
1	53.5	98.8	623	184	930	11966	7.57	2.34 ± 0.11
2	65.9	97.6	190	57	511	3783	12.8	4.49 ± 0.20
3	90.7	99.1	256	87	1246	5721	16.3	6.04 ± 0.27
4	49.7	95.7	755	616	3794	40014	12.1-	4.06 ± 0.18
5	87.4	96.1	423	213	2550	14017	14.7	5.36 ± 0.24
6	59.5	93.9	116	432	2746	28224	9.47	3.44 ± 0.15
7	87.9	97.6	105	128	1691	8383	15.1-	5.79 ± 0.26
8	67.3	96.9	685	118	1192	7774	16.1	5.63 ± 0.25
9	96.0	94.1	217	50	522	3275	11.0	3.79 ± 0.20
10	52.5	95.4	2199	630	2596	40932	7.77	2.66 ± 0.12
11	59.2	100.0	314	368	2308	23821	9.02	3.40 ± 0.15
12	77.5	99.7	404	226	2272	14703	12.9	4.73 ± 0.21
13	71.6	95.8	130	131	1118	8784	11.1	4.22 ± 0.19
14	77.8	96.6	1404	280	2264	18239	12.1	4.35 ± 0.20
15	66.4	95.3	263	563	5976	37094	16.9	5.95 ± 0.27
16	73.5	100.0	1608	1185	9309	76775	15.7	5.57 ± 0.25
17	90.0	90.6	234	58	1447	3796	32.9	12.9 ± 0.55
18	40.7	98.0	177	622	5277	41124	21.5	7.14 ± 0.32
19	71.1	94.4	250	v45	569	2971	18.7	6.25 ± 0.28
20	78.8	96.4	353	83	1404	5360	23.6	8.05 ± 0.36

TABLE 6 (con'd) - CALCULATION SHEET FOR DETERMINATION OF YTTRIUM-90 ACTIVITY IN MILK SAMPLES.

Sample No	Final Yield of Sr (%)	Final Yield of Y (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected initial Y^{90} activity of sample (uuc)	Activity of sample (uuc/gm Ca)
			t_1	(t_2-t_1)				
21	86.5	94.5	240	109	1908	6847	22.4	7.44 ± 0.33
22	71.1	97.9	244	75	1045	4738	20.3	7.75 ± 0.35
23	73.0	96.0	1241	124	1923	7975	34.0	11.3 ± 0.51
24	71.6	93.0	154	144	2268	9340	31.1	11.0 ± 0.50
25	45.4	97.2	642	726	6693	47119	22.0	7.48 ± 0.34
26	76.0	91.3	303	160	1951	10452	17.8	6.79 ± 0.30
27	76.8	99.5	1892	560	3931	30075	14.2	5.20 ± 0.22
28	42.2	96.9	801	658	3295	42755	10.4	3.87 ± 0.19
29	83.5	81.8	1410	151	1080	10042	11.2	4.15 ± 0.18
30	95.0	100.0	538	214	2327	13736	11.8	3.82 ± 0.16
31	71.0	94.2	1217	176	1428	11698	13.1	4.96 ± 0.23
32	80.0	97.7	696	77	1026	4921	18.8	6.52 ± 0.28
33	98.0	48.9	512	184	1766	12002	20.0	6.97 ± 0.31
34	95.0	98.6	973	144	3275	9478	29.7	9.77 ± 0.43
35	91.5	98.0	440	258	6160	17829	30.2	9.93 ± 0.46
36	30.0	64.8	841	735	8417	48007	68.3	21.9 ± 1.0
37	22.5	99.7	755	2130	23976	139813	64.5	20.2 ± 0.9
38	92.5	92.4	220	70	3033	4519	55.0	19.2 ± 0.9
39	88.0	97.2	292	266	10915	17351	54.9	19.3 ± 0.9

TABLE 7 - CALCULATION SHEET FOR DETERMINATION OF YTTRIUM-90 ACTIVITY IN RAINWATER SAMPLES (Straight Funnel Collector)

Sample No	Final Yield of Sr (%)	Final Yield of Y (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected initial Y^{90} counting rate (c/m)	Activity of sample (uuc) per lire
			t_1	(t_2-t_1)				
1	64.9	95.0	2819	188	617	12336	3.26	19.0 ± 2.4
2	55.4	97.9	115	227	2457	15291	21.0	22.3 ± 0.9
3	94.5	95.6	334	660	4799	43353	6.71	22.3 ± 0.9
4	91.2	97.0	363	72	666	4778	8.92	24.0 ± 1.4
5	95.5	99.5	414	586	31878	38063	59.2	15.0 ± 0.5
6	89.3	99.0	255	157	7887	9979	58.2	14.7 ± 0.5
7	91.7	96.3	301	61	315	3884	3.94	13.6 ± 1.4
8	92.2	97.7	1400	152	711	9806	4.06	15.0 ± 1.2
9	94.0	100.0	1538	68	764	4442	13.2	11.9 ± 0.6
10	96.8	94.2	1405	164	3317	10616	28.2	8.53 ± 0.39
11	84.6	97.4	352	137	3588	8781	32.0	9.60 ± 0.34
12	86.8	99.2	140	105	2948	6960	31.6	16.8 ± 0.6
13	62.0	98.0	1378	371	6425	24363	33.7	16.4 ± 0.5
14	88.6	96.3	718	540	4781	35396	9.75	44.4 ± 1.6
15	94.7	94.2	1261	372	3206	24545	9.82	46.4 ± 1.8
16	87.0	97.7	561	116	3847	7863	41.0	21.4 ± 0.7
17	98.0	39.7	4321	244	1998	16266	35.7	18.2 ± 0.8
18	84.9	91.4	1465	177	5242	11683	47.3	25.8 ± 0.9
19	97.0	96.9	3693	623	13122	40675	41.9	22.0 ± 0.7

TABLE 8 - CALCULATION SHEET FOR DETERMINATION OF YTTRIUM-90 ACTIVITY IN RAINWATER SAMPLES (Ion-Exchange Collector)

Sample No	Final Yield of Sr (%)	Final Yield of Y (%)	Counting times (min)		Total Geiger Rube Count	Total Guard Tube Count	Corrected initial Y^{90} activity (uuc)	Initial Y^{90} activity per litre (uuc)
			t_1	(t_2-t_1)				
1	28.2	100.0	139	41	469	2623	38.7	40.7 ± 2.4
2	32.9	94.2	423	2478	14263	164007	18.7	18.7 ± 0.8
3	93.4	96.0	153	266	1859	17446	7.67	19.7 ± 0.8
4	87.4	97.2	348	654	4092	42730	6.45	19.5 ± 0.9
5	52.0	93.0	137	166	4893	11042	66.4	16.7 ± 0.7
6	67.3	94.5	129	41	1746	2762	73.1	15.6 ± 0.6
7	82.6	95.4	157	85	248	5566	1.48	12.2 ± 2.5
8	35.0	88.9	699	122	564	7978	11.2	9.26 ± 0.80
9	41.0	92.7	825	519	2881	33597	13.0	12.6 ± 0.6
10	56.3	95.3	313	1360	10235	87664	33.4	10.1 ± 0.3
11	65.5	97.6	1553	1236	21714	80523	40.4	10.3 ± 0.4
12	90.7	91.9	365	275	4488	18004	21.1	15.5 ± 0.6
13	56.0	98.6	214	254	1515	16431	3.53	27.6 ± 2.2

TABLE 9 - CALCULATION-SHEET FOR DETERMINATION OF YTTRIUM-90 ACTIVITY IN TAPWATER SAMPLES

Sample No	Final Yield of Sr (%)	Final Yield of Y (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected initial Y^{90} counting rate	Initial Y^{90} activity per litre (u u c)
			t_1	(t_2-t_1)				
1*	91.6	99.0	270	209	662	13828	1.50	0.25 ± 0.03
2*	89.2	97.2	126	561	1702	36636	1.39	0.22 ± 0.02
3	83.0	95.0	1568	130	1695	8575	19.0	1.72 ± 0.07
4	68.1	75.0	150	114	1004	7744	14.1	1.88 ± 0.09
5	33.0	95.0	266	428	2429	28834	13.0	1.74 ± 0.08
6	58.0	99.7	1651	166	955	10873	9.14	1.22 ± 0.07
7	98.0	98.8	660	988	8969	63794	9.13	1.22 ± 0.04
8	94.5	57.0	294	295	2821	19696	15.4	2.05 ± 0.09
9	35.3	97.0	298	105	786	6815	17.4	2.31 ± 0.12
10	99.0	96.9	318	101	1550	6717	15.0	2.00 ± 0.09
11	78.8	96.6	435	91	1112	5919	14.7	1.95 ± 0.09
12	90.1	99.5	235	168	2026	10753	12.3	1.64 ± 0.06
13	98.5	85.0	280	51	576	3360	11.8	1.57 ± 0.08
14	60.3	96.0	235	92	823	6077	11.7	1.56 ± 0.08
15	80.0	100.0	1381	156	1481	10080	12.4	1.65 ± 0.08
16	52.2	98.0	283	172	1371	11069	12.7	1.69 ± 0.08
17	85.0	45.3	699	670	4247	43504	13.9	1.86 ± 0.09
18	44.8	98.9	220	1430	9649	92495	15.4	2.05 ± 0.09
19	61.7	96.0	333	419	4287	27217	15.6	2.07 ± 0.09
20	48.1	100.0	373	466	4051	30441	15.8	2.12 ± 0.10
21	75.6	99.5	592	686	7336	45265	13.9	1.87 ± 0.08

*Ice samples.

TABLE 10 - CALCULATION SHEET FOR DETERMINATION OF STRONTIUM- ACTIVITY IN MILK SAMPLES

Sample No.	Date of reception of sample	Weight of ash (gm)	Calcium Content of ash		Final Yield of Sr (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Activity of sample (c/m) at time of analysis
			%	gm		t_1	(t_2-t_1)			
1	Jan. 7, 1961	20.6	15.7	3.24	56.0	283	142	769	9266	6.31
2	Feb. 6,	19.0	15.0	2.85	69.0	167	45	473	3000	12.5
3	Feb. 20,	19.4	13.9	2.70	95.0	543	305	4295	20219	12.9
4	Feb. 28,	19.4	15.4	2.98	52.0	138	241	1834	15740	9.87
5	Mar. 6,	17.2	15.9	2.74	91.5	52	320	3732	20429	8.92
6	Mar. 13,	19.5	14.1	2.75	62.0	1272	187	1272	12144	7.93
7	Mar. 20,	19.2	13.6	2.61	92.0	150	109	887	7325	6.80
8	Mar. 27,	20.0	14.3	2.86	70.5	138	236	2197	15533	10.6
9	Apr. 3,	19.5	14.9	2.90	98.0	107	417	4275	30647	8.78
10	Apr. 10,	19.0	15.4	2.92	56.0	37	159	914	10520	6.83
11	Apr. 17,	18.3	14.5	2.63	62.0	108	534	3734	35777	8.24
12	Apr. 24,	19.5	14.0	2.73	80.5	57	338	3012	22331	8.74
13	May 1,	18.8	14.0	2.63	75.0	192	1094	8223	72634	7.52
14	May 8,	18.9	14.7	2.78	81.5	182	53	405	3448	9.68
15	May 15,	18.7	15.2	2.84	69.5	1141	66	724	4351	13.1
16	May 22,	18.3	15.4	2.82	75.0	148	90	855	6022	10.2
17	May 29,	19.3	13.3	2.55	94.2	98	351	3618	22923	9.13
18	June 5,	19.7	15.3	3.01	45.0	134	556	3842	24106	11.2
19	June 12,	18.6	16.1	2.99	74.5	44	75	750	4910	10.9
20	June 24,	18.8	15.6	2.93	82.5	90	233	4634	21270	6.80

TABLE 10 - CALCULATION SHEET FOR DETERMINATION OF STRONTIUM- ACTIVITY IN MILK SAMPLES

Sample No.	Date of reception of sample	Weight of ash (gm)	Calcium Content of ash		Final Yield of Sr (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Activity of sample (c/m) at time of analysis
			%	gm		t_1	(t_2-t_1)			
21	Aug. 2, 1961	19.7	15.3	3.01	86.5	112	87	1277	5663	6.67
22	Aug. 7,	18.6	14.1	2.62	74.5	125	293	3242	19453	5.54
23	Aug. 14,	19.5	15.4	3.00	75.0	57	406	6441	27049	8.42
24	Aug. 21,	19.3	14.7	2.84	75.0	184	64	806	4309	6.44
25	Aug. 28,	19.7	14.9	2.94	46.5	66	183	2450	11988	24.8
26	Sept 11,	19.0	13.8	2.62	76.5	57	30	452	1998	17.3
27	Sept.25,	19.2	14.0	2.69	80.0	108	251	2478	16449	10.0
28	Nov. 20,	19.5	14.0	2.73	47.5	95	270	1381	17872	10.8
29	Nov. 27,	19.7	13.7	2.70	87.0	67	96	848	6334	7.97
30	Jan. 15,1962	19.2	16.1	3.09	98.0	478	119	1130	7619	7.58
31	Jan. 22,	19.3	13.7	2.64	82.4	219	46	406	2953	8.45
32	July 3,1962	19.1	15.1	2.88	84.0	50	77	3653	5167	54.2
33	July 3,	19.0	15.1	2.87	99.0	303	114	6652	7413	57.7
34	July, 9,	18.7	16.3	3.04	98.0	102	150	15662	9750	103
35	July 9,	18.7	16.3	3.04	93.0	71	132	11748	8510	93.2
36	July 16,	18.8	16.6	3.12	32.0	100	79	5148	5161	211
37	July 16,	19.3	16.6	3.20	24.3	99	122	6596	7853	216
38	July 31,	19.0	15.1	2.87	98.0	106	30	4841	1999	165
39	July 31,	18.9	15.1	2.85	89.0	174	63	8282	4167	146

TABLE 11 - CALCULATION SHEET FOR DETERMINATION OF STRONTIUM-89 ACTIVITY IN RAINWATER SAMPLES (Straight Funnel Collector)

Sample No.	Time of Collection	Volume of Sample (ml)	Final Yield of Sr (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected initial activity of sample (uuc)	Initial Sr^{89} activity per litre (uuc)
				t_1	(t_2-t_1)				
1	May 8 - May 17	191	68.0	246	759	16406	50734	13.1	180 ± 32
2	May 17 - May 24	1050	58.0	125	70	5732	4563	62.3	150 ± 27
3	May 24 - May 31	334	99.0	125	1037	56131	68982	23.8	190 ± 33
4	May 24 - May 31	414	95.5	121	157	9750	10271	28.5	180 ± 32
5	May 31 - June 7	4400	100.0	66	32	13850	2183	194.8	110 ± 21
6	May 31 - June 7	4400	93.5	98	99	42576	6652	207.0	120 ± 22
7	June 7 - June 14	323	96.0	63	269	6498	17788	10.5	80 ± 14
8	June 7 - June 14	301	96.5	72	65	1841	,4472	12.4	110 ± 19
9	June 21 - June 28	1240	98.5	66	20	1531	1299	34.2	69 ± 12
10	June 28 - July 5	3680	96.5	84	91	12370	5970	62.8	42 ± 8
11	June 28 - July 5	3710	96.5	130	120	17057	7914	65.7	43 ± 8
12	July 5 - July 12	2090	90.5	87	96	13580	6301	69.7	82 ± 15
13	July 5 - July 12	2290	64.0	116	183	18621	11816	70.5	75 ± 13
14	July 12 - July 19	243	92.3	115	115	4543	7419	18.4	180 ± 32
15	July 12 - July 19	235	96.0	562	46	2068	3106	20.3	210 ± 37
16	July 19 - July 26	2130	92.5	122	52	9652	3481	89.7	100 ± 18
17	July 19 - July 26	2180	98.0	233	119	22998	7765	88.3	100 ± 18
18	July 19 - July 26	2040	87.3	85	61	9357	3890	78.4	89 ± 16
19	July 19 - July 26	2120	97.0	46	185	36231	11982	90.6	100 ± 18

TABLE - 12 - CALCULATION SHEET FOR DETERMINATION OF STRONTIUM-89 ACTIVITY IN RAINWATER (Ion-Exchange Apparatus)

Sample No.	Time of Collection	Volume of Sample (ml)	Final Yield of Sr (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected initial activity of sample (uuc)	Initial Sr^{89} activity per litre (uuc)
				t_1	(t_2-t_1)				
1	May 17 - May 24	950	29.5	204-	240	9167	15596	55.6	130 ± 24
2	May 17 - May 24	1000	34.5	67	52	2316	3447	55.9	140 ± 26
3	May 24 - May 31	390	98.0	149	295	13715	19394	20.5	130 ± 24
4	May 24 - May 31	330	91.5	314	334	13066	21708	18.4	140 ± 26
5	May 31 - June 7	3980	54.5	92	26	5194	1669	164.1	100 ± 19
6	May 31 - June 7	4700	70.5	69	228	53266	15073	149.2	78 ± 14
7	June 7 - June 14	121	86.5	146	104	775	6909	2.90	58 ± 11
8	June 21 - June 28	1210	35.0	46	293	6114	18926	24.4	50 ± 9
9	June 21 - June 28	1030	41.0	42	1213	32306	79242	27.2	70 ± 12
10	June 28 - July 5	3310	75.0	222	259	22420	17136	51.0	36 ± 5
11	June 28 - July 5	3930	96.0	168	147	20149	9606	63.6	38 ± 7
12	July 5 - July 12	1360	95.0	81	233	10810	15346	21.1	32 ± 6
13.	July 12 - July 19	128	59.5	66	91	1679	6093	12.7	90 ± 18

TABLE 13 - CALCULATION SHEET FOR DETERMINATION OF STRONTIUM-89 ACTIVITY IN TAPWATER SAMPLES

Sample No.	Time of Collection	Volume of Sample (ml)	Final Yield of Sr (%)	Counting times (min)		Total Geiger Tube Count	Total Guard Tube Count	Corrected Initial Activity of Sample (uuc)	Initial Sr^{89} Activity per litre (uuc)
				t_1	(t_2-t_1)				
1*	Apr. 20, 1961	6790	95.5	1096	492	1665	39089	0.70	0.09 ± 0.02
2*	Apr. 20, 1961	6930	93.0	141	160	675	10624	1.12	0.28 ± 0.05
3	May 21, 1962	12300	86.5	256	649	43826	43418	33.7	6.5 ± 1.2
4	May 27,	8350	71.0	128	287	10893	18771	23.0	6.3 ± 1.1
5	May 27,	8350	35.0	112	455	10594	29248	27.6	8.0 ± 1.4
6	June 3,	8350	60.5	268	96	2359	6419	16.9	4.8 ± 0.9
7	June 3,	8350	98.0	206	226	8783	14742	17.0	4.8 ± 0.9
8	June 11,	8350	98.5	94	62	2260	4049	15.8	3.7 ± 0.7
9	June 11,	8350	36.8	169	70	1127	4491	17.4	3.8 ± 0.7
10	June 18,	8350	99.0	190	327	12632	21794	16.8	4.1 ± 0.7
11	June 18,	8350	82.5	108	230	9571	15164	18.5	4.7 ± 0.9
12	June 26,	8350	92.0	113	51	1247	3361	11.1	2.5 ± 0.5
13	June 26,	8350	99.5	383	141	5049	9394	15.4	4.0 ± 0.7
14	July 3,	8350	61.0	216	330	7095	21723	14.5	3.7 ± 0.7
15	July 3,	8350	80.0	298	217	6441	14195	15.6	4.0 ± 0.7
16	July 10,	8350	52.5	290	280	6054	19540	17.0	4.4 ± 0.8
17	July 10,	8350	85.0	124	64	2460	4203	19.4	5.1 ± 0.9
18	July 4,	8350	44.8	81	24	425	1585	16.4	3.9 ± 0.7
19	July 4,	8350	64.5	162	129	3364	8535	17.0	4.1 ± 0.7
20	July 18,	8350	55.2	66	114	2481	7390	16.2	3.8 ± 0.7
21	July 18,	8350	85.0	485	634	21164	40828	16.7	4.2 ± 0.8

*Ice samples.

the appropriate equations developed in Chapter III, and for the previously obtained counting efficiency of 40.5%.

Table 10 is taken to illustrate the strontium analysis of the milk samples. In this table, the following points should be noted:

(1) The second column gives the time the liquid milk sample was received; at this time the milk was approximately one day old.

(2) The fifth column gives the computed weight of calcium used in the analysis; two independent and identical measurements of the calcium content were made with the average being reported.

(3) The sixth column gives the strontium yield on the basis of 20 mg of strontium carrier added.

(4) The seventh and eighth columns give the sample counting times; t_1 represents the time counting began and t_2 the time counting ended, both are measured from the time of precipitation of ferric hydroxide in the purification procedure.

(5) The ninth column gives the total geiger tube count which now represents disintegrations in the samples from both strontium-90 and strontium-89.

(6) The final column gives the strontium activity of the samples in counts per minute. The portion of the total activity due to strontium-89 can be computed by subtracting the contribution due to strontium-90. Strontium-89 activities in most of the samples listed in this table were very low at the time of analysis since about a year had elapsed from the date of collection to the time of counting. It was felt that strontium-89 activities could not be computed with any satisfactory degree of certainty. However, for the rainwater and tapwater samples strontium-89 activity has been determined. The strontium-90 portion of each sample was found by multiplying its subsequently found total yttrium-90 activity by

28.9%, that is, the strontium-90 counting efficiency. Final strontium-89 activities were found by employing the estimated 35% strontium-89 counting efficiency. No corrections have been made for the build up of yttrium-90 before and during the counting of strontium-89 and strontium-90. This correction, if made, would have been well within the assumed 17% error of the strontium-89 counting efficiency.

Error quoted for the final results were computed by combining the standard deviation of the total count, $(N)^{\frac{1}{2}}$, directly with the estimated chemical errors. When possible at least 3000 counts were registered.

With very minor differences the remaining tables in this series are identical with those described above. It is to be noted that for most of the collected samples two independent and identical measurements were made.

The final results of strontium-90 activities in milk are summarized in Table 14. The last four samples in the table are renumbered since each sample represents the average of two independantly determined values. In Tables 15 and 16 the data for rainwater and tapwater are given respectively. Strontium-89 and strontium-90 activities were computed for most of the samples. Each value represents the average of two independant results, thus the sample numbers are different from those in the calculation sheets. For rainwater samples, results are given conjointly for the two rain collecting units. It is noted that both sets of results show very good agreement. In some cases, however, the straight funnel collector gives results that are slightly greater than the corresponding ones for the ion-exchange collector. This slight increase is attributed to the fact that the straight funnel collector is open to the atmosphere and evaporation can

TABLE 14

STRONTIUM-90 ACTIVITIES IN MILK

Sample No.	Time of Collection	Sr^{90} activity per gm calcium (uuc)	Sample No.	Time of Collection	Sr^{90} activity per gm calcium (uuc)
1	Jan. 7, 1962	2.42 ± 0.11	19	June 12,	6.40 ± 0.29
2	Feb. 6,	4.63 ± 0.21	20	July 24,	8.22 ± 0.37
3	Feb. 20,	6.22 ± 0.28	21	Aug. 2,	7.60 ± 0.34
4	Feb. 28,	4.25 ± 0.19	22	Aug. 7,	7.91 ± 0.36
5	Mar. 6,	5.52 ± 0.25	23	Aug. 14,	11.5 ± 0.52
6	Mar. 13,	3.60 ± 0.16	24	Aug. 21,	11.2 ± 0.51
7	Mar. 20,	5.96 ± 0.27	25	Aug. 28,	7.61 ± 0.35
8	Mar. 27,	5.80 ± 0.26	26	Sept 11,	6.93 ± 0.31
9	Apr. 3,	3.90 ± 0.21	27	Sept 25,	5.29 ± 0.22
10	Apr. 10,	2.80 ± 0.12	28	Nov. 20,	3.94 ± 0.19
11	Apr. 17,	3.49 ± 0.16	29	Nov. 27,	4.21 ± 0.18
12	Apr. 24,	4.86 ± 0.22	30	Jan. 15, 1962	3.87 ± 0.16
13	May 1,	4.33 ± 0.20	31	Jan. 22,	5.01 ± 0.23
14	May 8,	4.47 ± 0.21	32	July 3,	6.75 ± 0.20
15	May 15,	6.10 ± 0.28	33	July 9,	9.85 ± 0.31
16	May 22,	5.72 ± 0.26	34	July 16,	21.1 ± 0.7
17	May 29,	13.1 ± 0.56	35	July 31,	19.3 ± 0.6
18	June 5,	7.35 ± 0.33			

TABLE 15

STRONTIUM ACTIVITIES IN RAINWATER

Sample No.	Time of Collection	Ion-exchange collector		straight funnel collector	
		Average Sr^{90} activity per litre (uuc)	Average Sr^{89} activity per litre (uuc)	Average Sr^{90} activity per litre (uuc)	Average Sr^{89} activity per litre (uuc)
1	May 8 - May 17,1962	- - - -	- - - -	19.0 ± 2.4	180 ± 32
2	May 17 - May 24,	18.7 ± 0.8	140 ± 18	22.3 ± 0.9	150 ± 27
3	May 24 - May 31,	19.6 ± 0.6	140 ± 18	23.2 ± 0.8	190 ± 23
4	May 31 - June 7,	16.2 ± 0.5	90 ± 12	14.9 ± 0.4	120 ± 15
5	June 7 - June 14,	12.2 ± 2.5	58 ± 11	14.3 ± 0.9	100 ± 12
6	June 14 - June 21,	(no rain)	- - - -	- - - - -	- - - -
7	June 21 - June 28,	11.0 ± 1.7	60 ± 8	11.9 ± 0.6	69 ± 12
8	June 28 - July 5,	10.2 ± 0.3	37 ± 5	9.07 ± 0.55	43 ± 6
9	July 5,- July 12,	15.5 ± 0.6	32 ± 6	16.6 ± 0.4	79 ± 10
10	July 12 - July 19,	27.6 ± 2.2	160 ± 30	45.4 ± 1.2	200 ± 25
11	July 19 - July 26,	- - - - -	- - - -	21.9 ± 1.0	97 ± 9

TABLE 16

STRONTIUM ACTIVITIES IN TAPWATER

Sample No.	Time of Collection	Average Sr^{90} activity per litre (uuc)	Average Sr^{89} activity per litre (uuc)
1	Apr. 20, 1961	0.25 ± 0.02	- - - -
2	May 21, 1962	1.72 ± 0.07	6.5 ± 1.2
3	May 27,	1.81 ± 0.06	7.2 ± 0.9
4	June 3,	1.22 ± 0.04	4.8 ± 0.7
5	June 11,	2.05 ± 0.09	3.8 ± 0.5
6	June 18,	1.98 ± 0.07	4.4 ± 0.6
7	June 26,	1.61 ± 0.05	4.0 ± 0.7
8	July 3,	1.61 ± 0.06	3.9 ± 0.5
9	July 10,	1.78 ± 0.07	4.8 ± 0.6
10	July 4,	2.06 ± 0.08	4.0 ± 0.5
11	July 18,	2.12 ± 0.10	4.0 ± 0.5

easily take place. On the other hand the reservoir for the ion-exchange collector is completely closed and consequently evaporation is more difficult.

The variation of strontium-90 activities in milk with time are shown in Figure 10. There was significant rise in activity during the period of investigation. The ratio between the lowest activity in 1961 to the highest in 1962 was found to be approximately 10, however, accurate estimates of this ratio cannot be made since the results also show a seasonal variation.

Strontium-90 levels in rainwater samples for the two different collectors are plotted in Figures 11 and 12. Strontium-89 levels in the same samples are shown in Figures 13 and 14. It can be seen that the variation of strontium-90 and strontium-89 levels are in good agreement. However, the statistical significance of these results is difficult to be found since the period of observation was short. It is hoped that future results will help in interpreting the present findings.

The data obtained from tapwater samples are plotted in Figures 15 and 16. In general, the results show more statistical fluctuations than the other collected samples. Strontium-90 activities show a very similar increase to that of milk samples over the period of the last year, although only two values were available in 1961. Variation in strontium-89 activities is somewhat different from those of strontium-90 unlike the rainwater samples. However, it is very difficult to reach any significant conclusion with so few data.

No comparison between the present results and the national or international findings by other investigators was made since other data over

Fig. 10 - Strontium-90 Activities in Milk

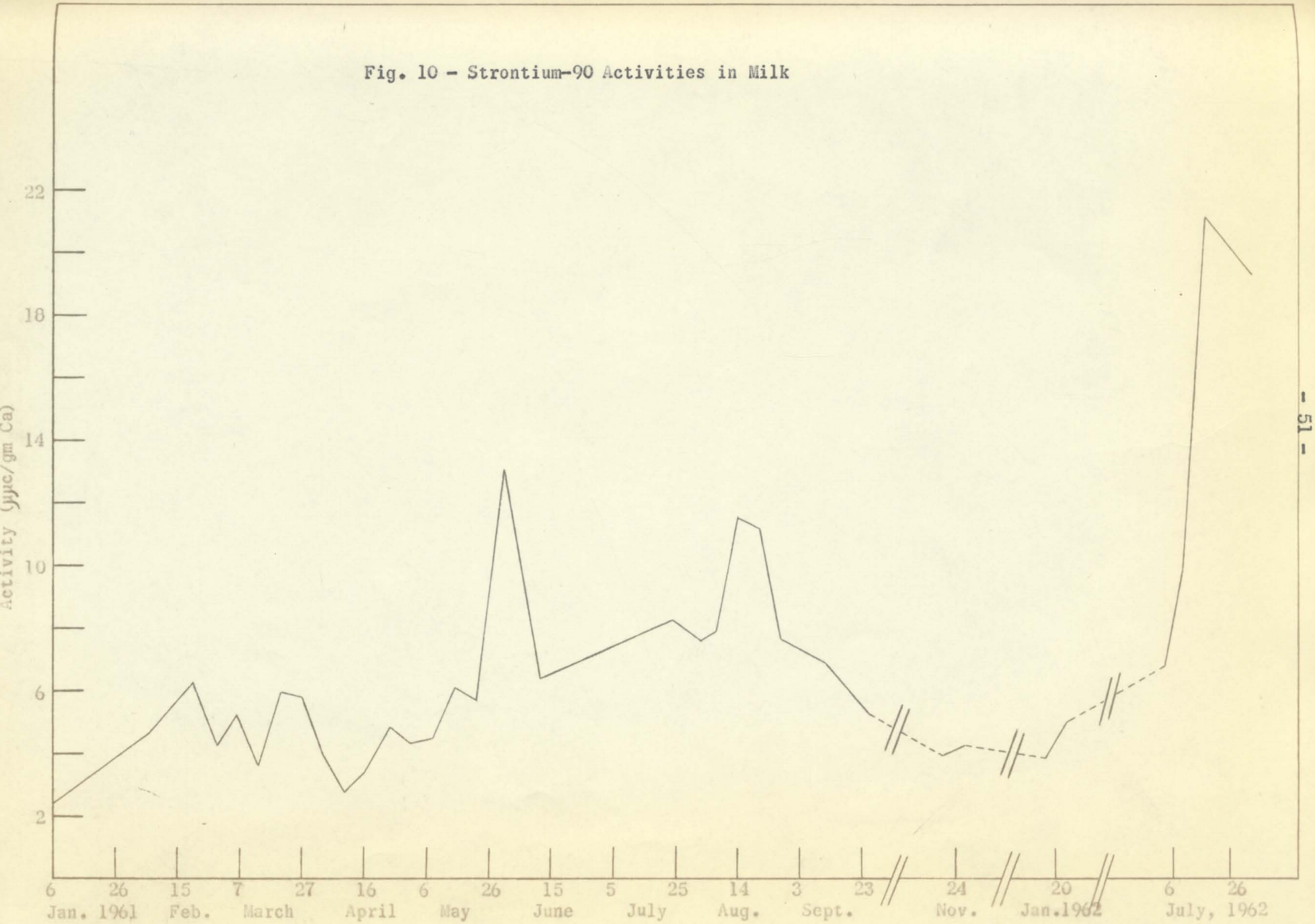


Fig. 11 - Strontium-90 Activities in Rainwater (Ion-exchange collector)

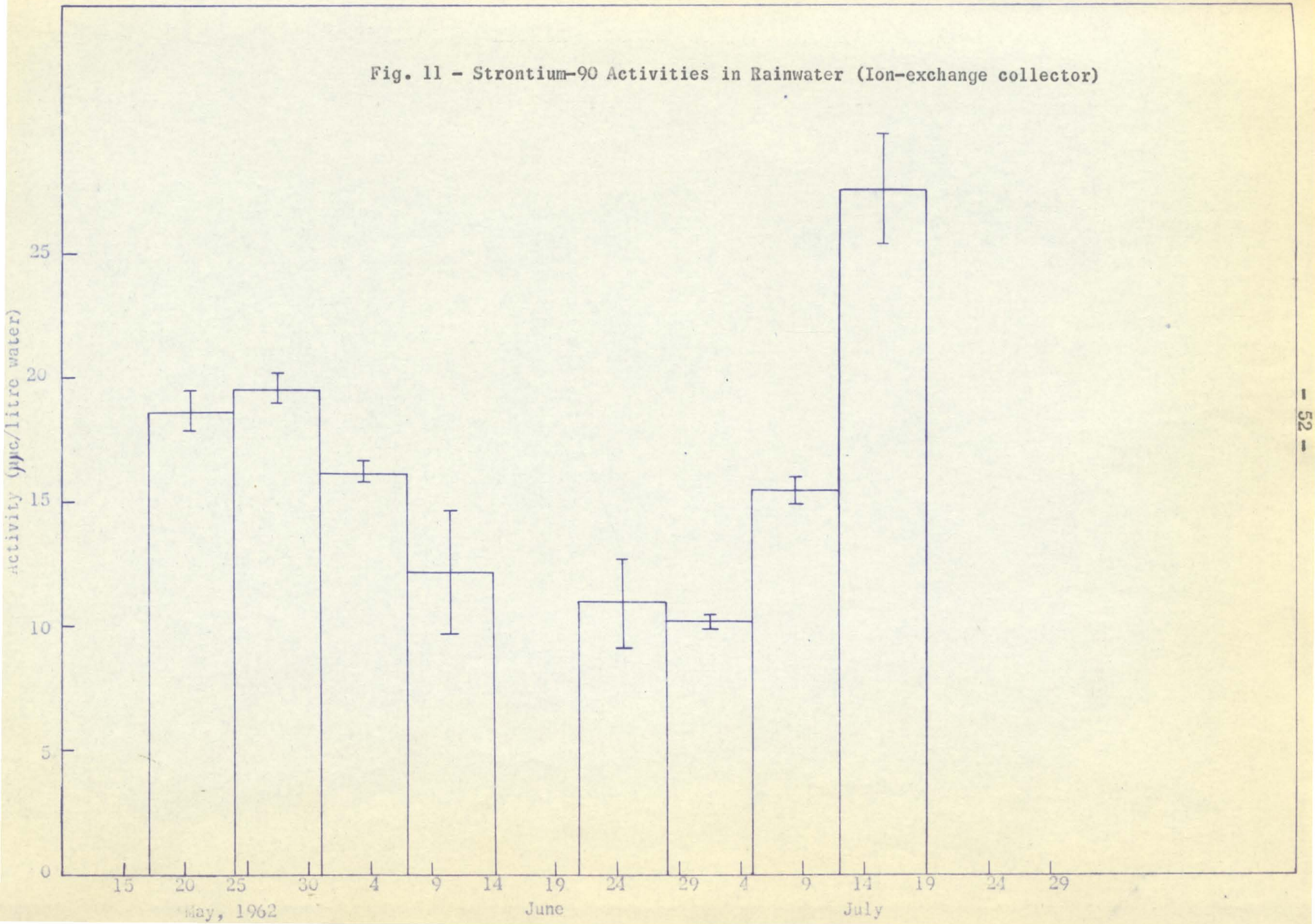


Fig. 12 - Strontium-90 Activities in Rainwater (Straight funnel)

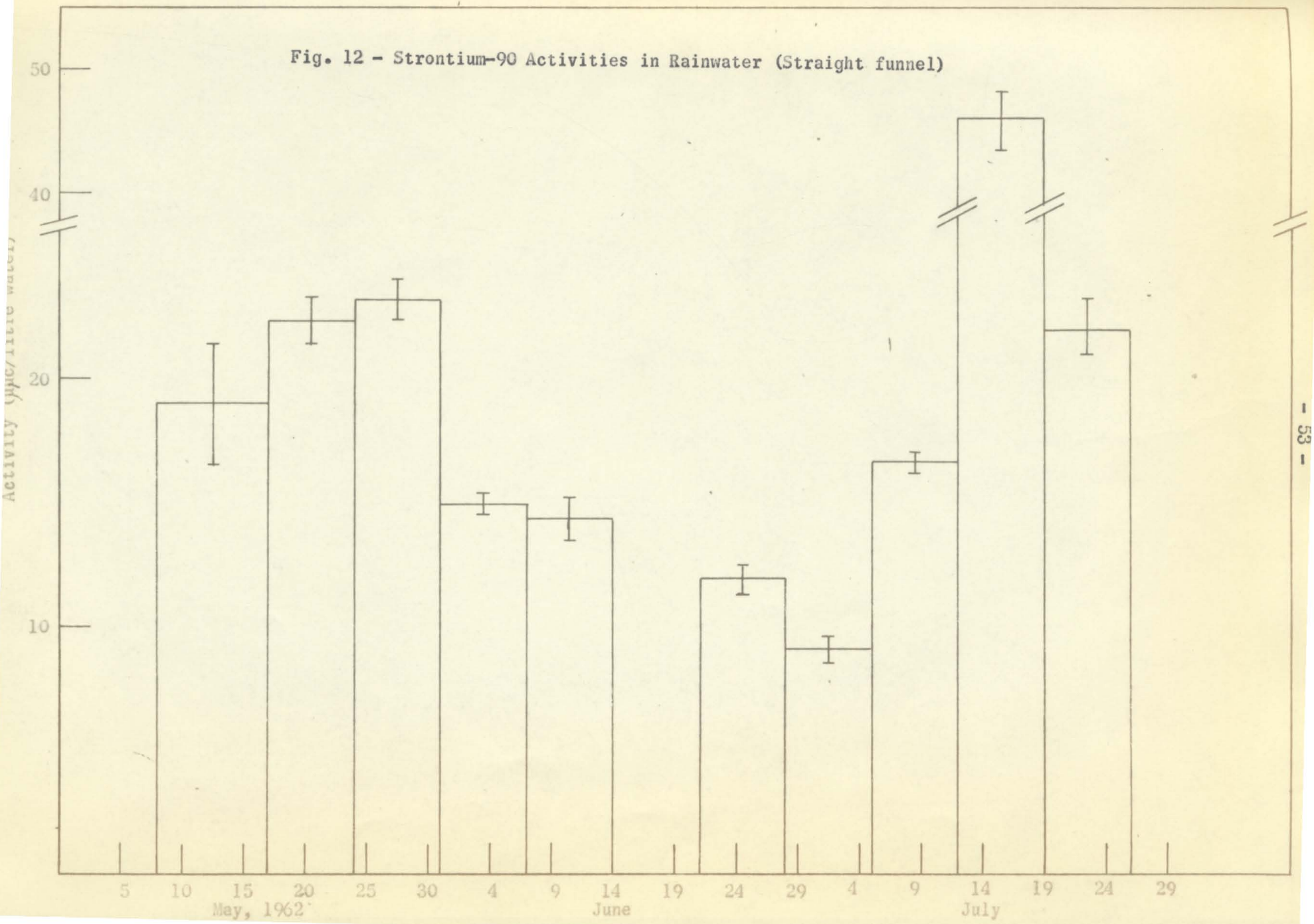


Fig. I3 - Strontium-89 Activities in Rainwater (Ion-exchange collector)

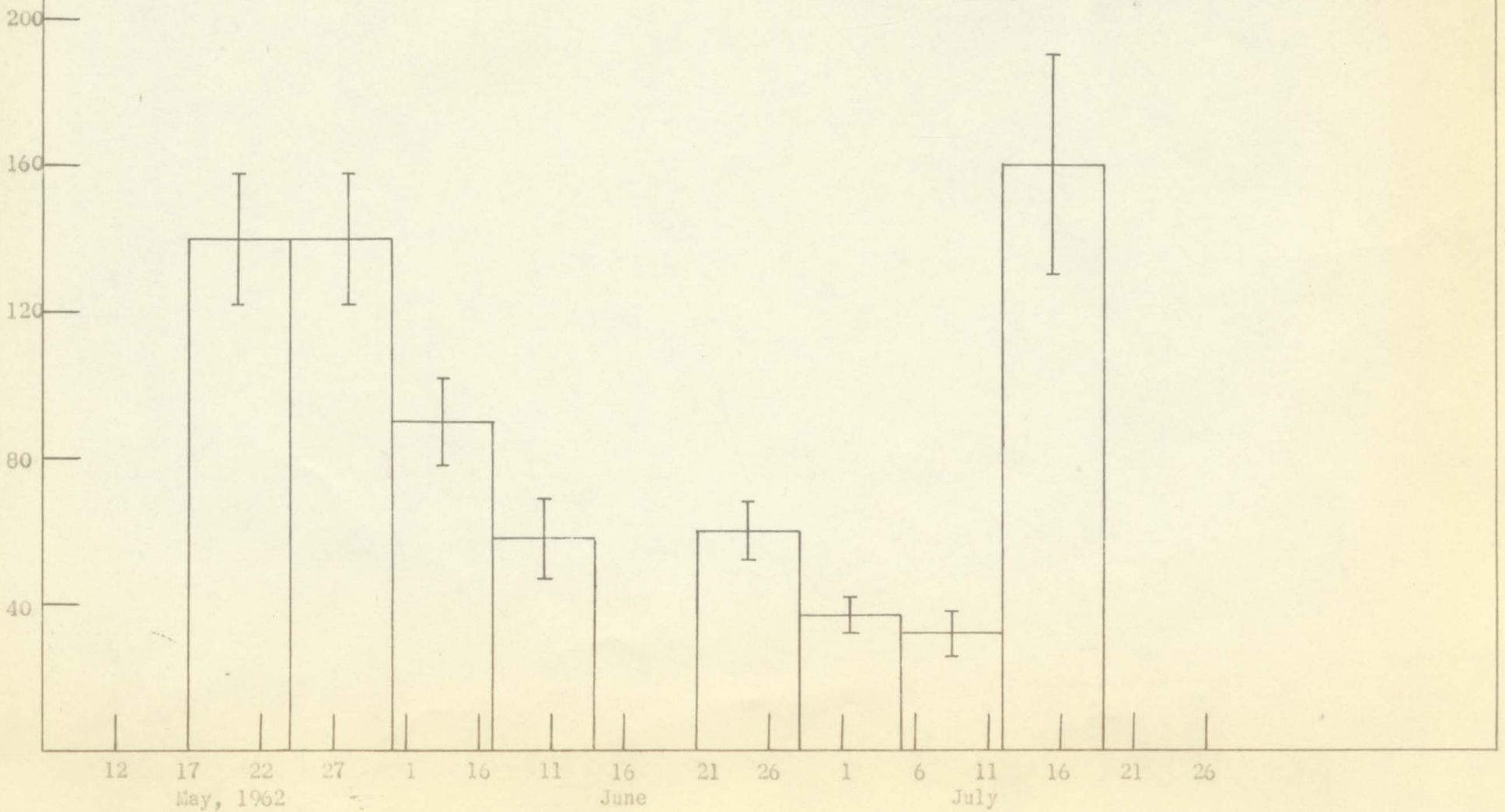
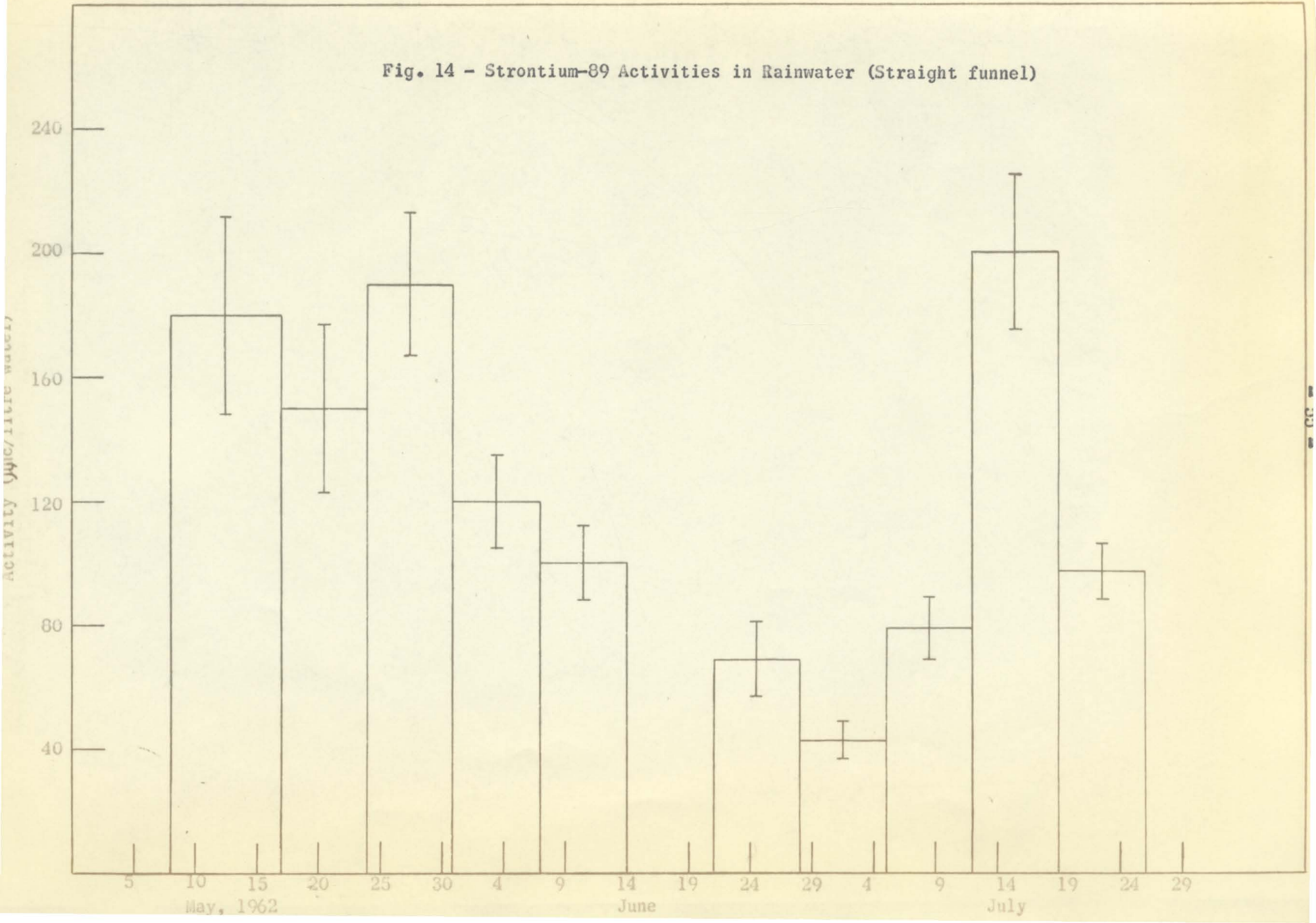


Fig. 14 - Strontium-89 Activities in Rainwater (Straight funnel)



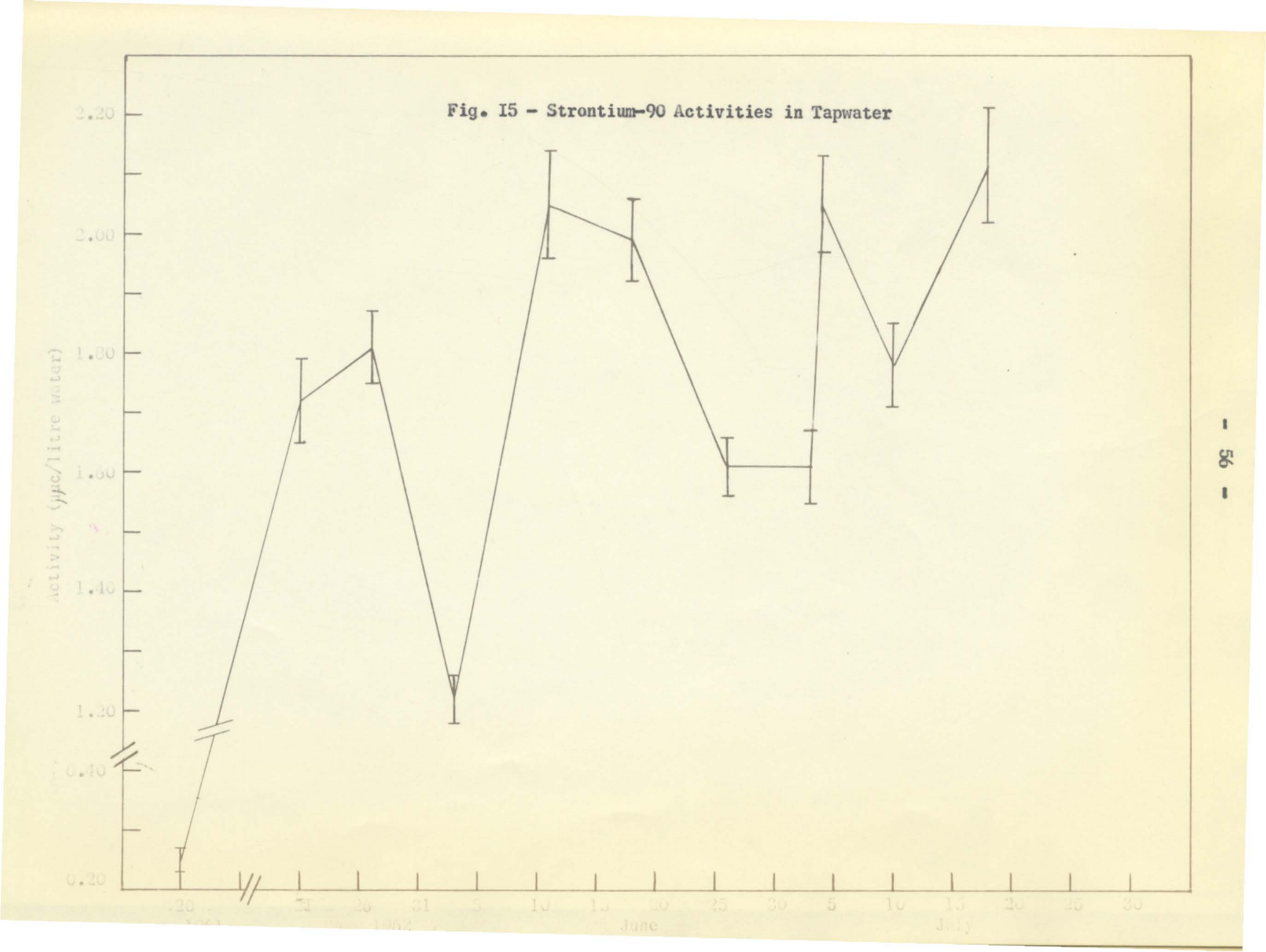


Fig. I5 - Strontium-90 Activities in Tapwater
Activity (μμc/litre water)
2.20
2.00
1.80
1.60
1.40
1.20
0.40
0.20
June
July

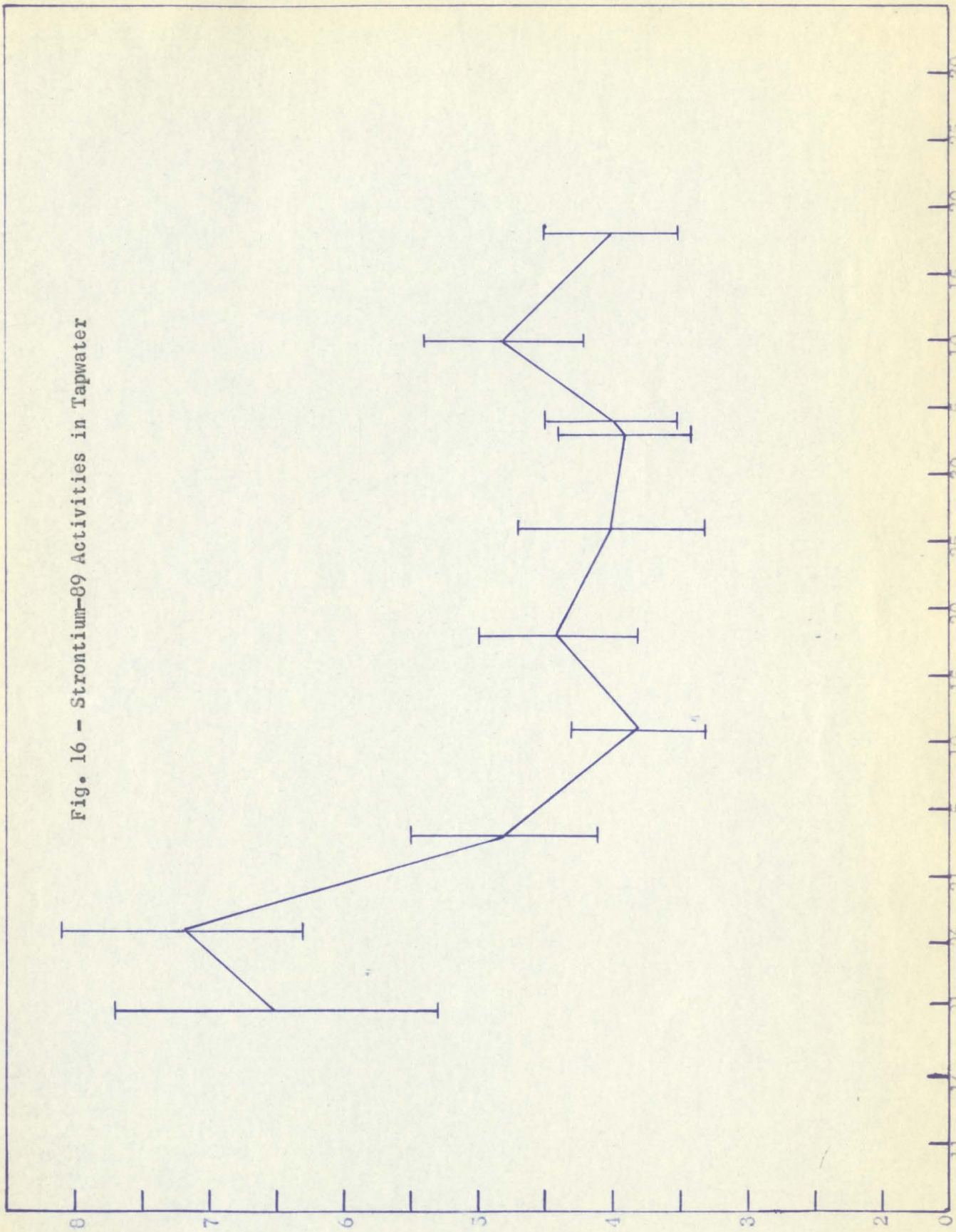


Fig. 16 - Strontium-89 Activities in Tapwater

the same period were unavailable when this thesis was prepared.

BIBLIOGRAPHY

1. Aler, B., Edvarson, K., Report to the United Nations, A/Ac. 82/R-52, April 17, 1957.

2. Anikina, M. P., Ivanov, R. N., Kukavadze, G. M., Ershler, B.V., Atomnaya Energ. 4, 198 (1958).

3. Atomic Energy of Canada Limited, National Department of Health and Welfare, Ottawa, CRC-688, Feb., 1957.

4. Brown, R.D., O'Donnell, T. A., Manuel of Elementary Practical Chemistry, The Macmillan Company, Toronto, 1958.

5. Coryell, C. D., Sugarman, N., Radiochemical Studies: The Fission Products, Book 3, McGraw-Hill Book Company, Inc., 1951.

6. Osmond, R. G., Owers, M. J., J. Energ. Nuclear Chem., 9, 96 (1959).

7. Osmond, R. G., Owers, M. J., Healy, C., Mead, A. P., AERE-R 2899, 1959.

8. Robert, J., J. Phys. Radium 20, 830 (1959).

9. United States Atomic Energy Commission, HASL-42, October, 1958.

10. Lange, N. A., Handbook of Chemistry, 10th Edit. McGraw-Hill Book Co., 1961.

ACKNOWLEDGEMENTS

The research described in this thesis was supervised by Professor C. W. Cho to whom the author is greatly indebted for his guidance both during the course of the work and in the preparation of the thesis.

The assistance and direction offered by Professor S. W. Breckon is also gratefully acknowledged.

The writer wishes to express his thanks to the Chemistry Department of Memorial University for the periodic use of their chemical supplies and equipment and to W. Pearce and R. Bishop for their assistance in the experimental work.

Financial assistance in the form of graduate fellowships from Memorial University for the years 1960-62 is also gratefully acknowledged.